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Determining the composition of post-consumer flexible multilayer plastic packaging with near-infrared spectroscopy

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ABSTRACT

Flexible multilayer plastic packaging (MPP) has grown in popularity in the last years especially in food and medical sectors, and its share in the packaging industry is expected to increase further. Compared to traditional packaging with same functionalities, MPP is characterized by lower energy consumption in production and a reduced packaging weight. So far, the recycling of post-industrial MPP with specific material composition has been achieved by several companies. To our knowledge, all existing MPP recycling processes require a known material combination. In contrast to post-industrial MPP, post-consumer MPP still ends up in incinerators or as low-quality products, mainly because of the lacking ability to sort. This study investigates the detectability of post-consumer MPP with near-infrared spectroscopy, the state-of-the-art technology for sensor-based waste sorting. Firstly, MPP classification with near-infrared spectroscopy was analyzed with clean samples. Subsequently, the effect of waste collection and preprocessing in sorting plants on MPP classification was investigated. For this purpose, clean samples were covered with water and oil and mixed with lightweight packaging waste in a drum sieve. The results show it is possible to classify post-consumer MPP based on near-infrared spectra according to different sorting strategies. For the existing recycling processes which are suitable for post-consumer MPP, the corresponding object-based classification accuracy was found to exceed 96%.

1. Introduction

Flexible multilayer plastic packaging (MPP) is a crucial packaging material that combines the best properties of different types of plastics in a layer-wise structure. The use of MPP has increased significantly in recent years and is expected to increase further in the future (Briassoulis et al., 2017). In 2018, Europe-wide plastics production amounted to 51.2 million Mg, with packaging accounting for the largest share (40 wt%) (Plasticeurope, 2019). Among all packaging materials produced, MPP films account for 17 wt% (Mumladze et al., 2018). MPP films are characterized by their functional, protective and decorative properties, which result from a combination of at least two thermoplastics. Due to these characteristics, MPP is mainly used in the food industry to extend the shelf life of products and at the same time reduce food waste, which currently accounts for one third of all food produced (Morris, 2017). Typical products of MPP include bags, pouches and shrink films.

MPP consists of individual layers of varying thickness, which are produced in a coextrusion or lamination process to form a homogeneous multilayer (Cinelli et al., 2016). MPP films are only a few micrometers thick, and the number of layers varies between three and twenty layers. Not all layers are made of plastics; adhesive layers, ink or metal layers can also be incorporated (Niaounakis, 2019). In general, MPP consists of several main layers (>10 μm), which are often connected by thin adhesive layers with a thickness of 1–3 μm (Mieth et al., 2016). As adhesive layers are not always necessary and have – compared to the main layers – a negligible thickness, they are not considered as individual layers in this study.

The simplest structure of MPP is a two-layer plastic composite. This consists of two main layers: an inner layer, which contacts the product and must be chemically inert, and an outer layer, which provides structural stability, abrasion resistance, surface printability and high temperature resistance to prevent melting during sealing (Morris, 2017). As the layer number increases, more combinations of different materials and thicknesses are possible. Frequently used materials in MPP are polyethylene (PE),

Abbreviations: EVOH, ethylene vinyl alcohol; MPP, multilayer plastic packaging; NIR, near-infrared; PA, polyamide; PE, polyethylene; PET, polyethylene terephthalate; PLS, partial least squares; PP, polypropylene.

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polypropylene (PP), polyethylene terephthalate (PET), polyamide (PA), ethylene vinyl alcohol (EVOH) as well as metallized layers for light barriers (Butler and Morris, 2016).

Currently, MPP is either thermally utilized (Kaiser et al., 2018), reproduced to low-quality products ("downcycling") (Plasticeurope, 2019) or landfilled (Kaiser et al., 2018). Furthermore, valuable mono-material fractions are contaminated by MPP due to incorrect classification and sorting of MPP as a pure material on the basis of the plastic type of the layer which is exposed to the sensor (Ragaert et al., 2017). To achieve a higher recycling rate and move further towards a circular economy, it is necessary to separate MPP from monolayer plastics and the plastics contained in MPP need to be recovered in a separate recycling process.

For post-industrial MPP waste with specific compositions, individual pilot rigs have been implemented and experimental plants are under construction. Saperatec GmbH from Germany has developed a process with physical delamination of MPP (Niaounakis, 2019) through reduction of interfacial tensions between adhesive layers and material layers by a liquid separating solvent (Kaiser et al., 2018). This process is limited to MPP with at least one layer of metal and one layer of plastic (Niaounakis, 2019). Fraunhofer IVV and Creacycle GmbH have developed the solvent-based separation process "CreaSolv" for the separation of MPP, which is suitable for both post-industrial and post-consumer MPP (Frauenhofer IVV, 2020). This process has been already implemented in pilot plants in Indonesia, but they are only able to process post-industrial waste in form of multilayer plastic bags and only PE is recovered (Unilever, 2020). A pilot plant in Germany is under construction with the goal of recycling PE and PP from MPP (Bundesministerium für Bildung und Forschung, 2018). Another process that has already been implemented on a large scale is the "Newcycling" process of APK GmbH. This solvent process is suitable for both post-industrial and post-consumer MPP (APK AG, 2020). A recycling plant in Germany has been in operation, which is exclusively suitable for post-industrial MPP consisting of PE and PA (Geigermedien GbR., 2018).

All available industrially implemented processes for recycling MPP have in common that the processes are only suitable for specific material combinations and the input material or the composition of MPP must be known. Thus, recycling post-consumer MPP waste requires the generation of a sorted MPP waste fraction with a known composition. For post-consumer MPP, there is still no industrial and economic solution, as it is not recognized in the sorting process because of the wide variety of materials used (Niaounakis, 2019).

In Germany, post-consumer plastic packaging waste is collected – together with other packaging material – as lightweight packaging (LWP) waste. To recover materials from LWP waste in a sorting plant, the material flow is firstly liberated with sack-opener and then mechanically pretreated for further sensor-based sorting (Feil and Pretz, 2020). For sensor-based sorting of plastic, the state-of-the-art technology is near-infrared (NIR) spectroscopy because of the high accuracy in sorting and the high throughput. NIR spectroscopy offers the possibility of obtaining spectral properties of an object, which contain information about the chemical composition and the physical state of the material (Linnemann, 2008). NIR-active materials, for example polymers, have characteristic spectra, on the basis of which the material types can be determined. Besides, surface condition (e. g. roughness and surface moisture) influences the spectra of plastics (Küppers et al., 2019). NIR-inactive materials, for example ceramic tiles, contrarily, reflect NIR light without absorption. The disadvantage of NIR technology in classifying plastics is the misclassification of black materials with carbon black as coloring agent, as they do not reflect sufficient light for analysis (Beel, 2017).

In this study, NIR spectroscopy is used to determine the detectability of MPP. As films are separated from three-dimensional waste in the recycling plant, MPP was analyzed through spectral comparison between different films. We analyzed spectra of different clean multilayer samples and investigated the potential for a qualitative and a quantitative determination of MPP composition. To further determine the feasibility of NIR-based sorting of MPP from post-consumer waste, we simulated different characteristics of post-consumer waste streams such as different material presentations to the sensor unit as well as different types of material contamination and studied their effects on the detection and classification of MPP.

2. Methods and material selection

To investigate the detectability, a test rig was arranged and the settings were adjusted for an appropriate detection of multilayer samples. For this purpose, an analysis program has been developed to classify samples through comparing and analyzing the spectra of different material combinations.

2.1. Capturing and analysis of NIR spectra

NIR spectra of the samples were captured with a Helios-G2-320 NIR sensor from EVK DI Kerschhaggl GmbH (Raaba, Austria) in a spectral range of approx. 930 to 1700 nm. The spectral resolution is 3.1 nm/pixel, and the spatial resolution is 1.25 mm/pixel. Four halogen lamps with a power of 400 W each were used as emitters. The reflection of radiation from the surface is captured by the NIR sensor, as shown in Fig. 1.

Usually in sorting plants, the background of a NIR-based sorter is a black conveyor belt. Black materials, as mentioned above, reflect less radiation, and can thus be easily distinguished from non-black objects. Therefore, spectra of all the samples were captured and analyzed with black conveyor belt as background. However, since most MPP materials are transparent and have low thickness (<150 μm), the intensity of the reflected radiation is relatively low. To clearly demonstrate the differences in spectra, a gray, NIR-inactive ceramic tile was used – in addition to conveyer belt – as background for specific demonstration purposes.

The captured spectra were analyzed with a self-developed analysis program. For analysis, the first derivative of original reflection is applied as it better shows the change in reflectance intensities. For classification and regression, a partial least squares (PLS) discriminant analysis algorithm was used. In both cases, a components number of 100 was chosen and the implementation was based on the function "PLSRegression" from Scikit-learn (Pedregosa et al., 2011). The samples were pixel-based analyzed. During the training process, an area of 40*40 pixels was selected

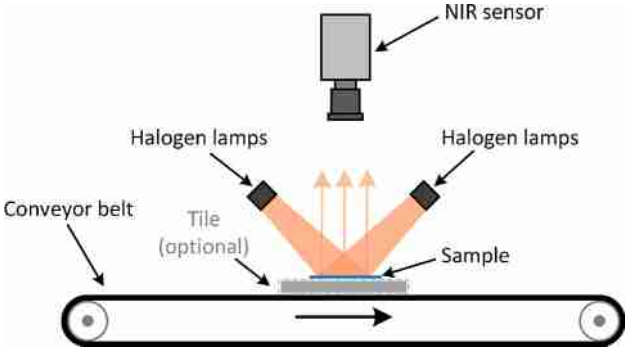

Fig. 1. NIR sensor and illumination setup.

for each class and divided into 70% training and 30% test data. To evaluate the model performance, each class contains the same number of instances (6000 pixels). For this purpose, a different area of 60*100 pixels from each sample was selected. In case n samples belong to the same class, 6000/n pixels from one sample are randomly selected, so that for each class in total 6000 pixels are analyzed. To further show the robustness of the achieved pixel-based prediction results, the test data with 6000 pixels was randomly divided into 10 subgroups with 600 instances each, and the 95% confidence interval is determined on the basis of the achieved results of all subgroups. The classification process is then evaluated on the basis of the metric accuracy which is the percentage of right classified instances to the overall number of instances in each group. For object-based classification, the area of 60*100 pixels is selected for each sample. If more than 70% of the pixels are classified to the right class, it is assumed that the sample is object-based classified to the right class. The regression process was evaluated with R^2-score which is the proportion of variation in the prediction that is explained by the spectrum. The range of R^2-score is usually [0, 1]; the closer R^2-score is to 1, the better is the prediction of the investigated model.

In summary, NIR spectra of samples are collected in 220 spectral points, and the 100 most crucial components are used for analysis. Different areas are chosen for model training and evaluation of the classification ability. For qualitative analysis, a pixel-based classification with PLS discriminant analysis was conducted, and PLS regression was used for quantitative analysis.

2.2. Material selection

To investigate the classification of MPP, 25 different monolayer and 95 different multilayer samples, which consists of the most often used plastic types (PE, PP, PET, PA, EVOH) in film-based plastic packaging, were analyzed. The used multilayer samples differ in the number of layers, the thickness of each layer and their appearance (transparent or printed). They were available in two, three and four layers and have a total thickness between 35 μm and 150 μm. In total, there were 16 different material combinations, as shown in Table 1.

The samples were virgin material and produced with coextrusion by the company Estiko-Plastar AS (Tartu, Estonia). Flexography was used for printing the samples, and polyurethane based two-component adhesive layers are used between two plastic layers for all samples. To fit the conveyer belt dimensions of the used test rig, the samples were cut to approx. DIN A4 format. The samples were named with material type and thickness in μm of each material and the side of the sample which is exposed to the sensor is listed first. For example, PET30/PP20/PE50 means a non-printed multilayer sample with a PET layer of 30 μm, a PP layer of 20 μm and a PE layer of 50 μm, and the PET layer was exposed to the sensor. A "met" after one material indicates a metalized layer, for example, PETmet means a metalized PET layer. If the sample is printed, a "P" is added behind the designation, e.g.: PET30/PP20/PE50P. The spectra of the materials with a tile as background are additionally marked with a "T" behind the designation. For EVOH-containing samples, all EVOH layers have a thickness of 5 μm, and they are designated as e.g. PE/EVOH/PE90, where 90 μm is the total thickness.

Table 1
Overview of material combinations of used multilayer plastic samples.

Two-layer samples	Three-layer samples	Four-layer samples
PE/PE	PE/EVOH/PE	PET/PE/EVOH/PE
PET/PE	PP/EVOH/PP	OPP/PE/EVOH/PE
PET/PP	PET/PETmet/PE	
OPA/PE	OPP/PETmet/PE	
OPA/PP	OPP/PETmet/PP	
OPP/PE		
OPP/PP		
OPP/OPP		
OPP/OPPmet		

OPA: oriented PA; OPP: oriented PP.

Samples were folded to simulate a bag packaging and different presentations of MPP in waste. As MPP is most often used in the food industry, the effects of impurities were determined with water and oil. Besides, the collection process of LWP waste in Germany and the preprocessing of waste in sorting plant (moisture adhesion and surface contamination) were simulated. The samples were placed respectively in water and oil baths for 12 h, followed by a mixing process with post-consumer LWP waste, which was collected from Remondis GmbH & Co. KG (Erftstadt, Germany), a LWP waste sorting plant. The mixing process was carried out with a drum sieve with a radius of 0.75 m, a length of 1 m and an angular velocity of 0.82 s^{-1} for 30 min. The used LWP waste material has a volume of 0.27 m^3. After mixing with LWP waste, the MPP samples were captured and analyzed with the aforementioned test rig.

3. Results and discussion

To determine the detectability, spectra of different samples were compared and analyzed. To illustrate the difference in spectra, we show the mean value of first derivatives of all spectra in a selected area with 40*40 pixels for each sample. For pixel-based classification, the accuracy is shown as mean accuracy ± 95% confidence interval.

3.1. Detectability

3.1.1. Non-detectable case

Among spectra of all samples, it was not possible to analyze and classify the spectral data for several cases. Some transparent PET12-containing two-layer samples have shown wave-like spectra in wavelength range of 1400–1700 nm. Besides, the form and peak positions of each pixel are different from those of other pixels. However, it was determined that if a PET12/PE multilayer sample is printed or the PE layer has a higher thickness (≥40 μm), the spectra can be analyzed and classified. This rests on the assumption that the wave-like spectra occur in non-printed samples with PET or PP layers in low thickness (e.g. PET12, PP15), and with increasing thickness and layer number, this problem ceased to exist. The possible reason for wave-like spectra is direct and non-dispersive reflection from the very glossy and highly reflective surface of thin layers. For a more detailed explanation of the reason for a similar phenomenon in wave-like spectra, further research is needed.

Spectra of overexposed pixels are another case which cannot be analyzed. This problem occurs at some areas of individual samples with bended surfaces. In this case, some pixels reflected too much light in the whole wavelength area which is beyond the measuring range of the sensor, such that the data could not be analyzed. The reason for overexposed pixels is the material presentation. If the presentation of sample is different, the position of overexposed area changes accordingly. For different samples and different material presentations, the overexposed area was always less than 10% of the total sample's area, which has a limited influence on object-based classification. For the analysis, pixels with an average intensity higher than 90% of the sensor measuring range after calibration are considered as overexposed pixels and these pixels were removed to not influence the mean spectrum and classification results.

3.1.2. Comparison of MPP spectra to background and monolayer

To determine the detectability, spectra of samples were firstly compared with background spectra. The comparison showed that all the samples have different spectra than the background and were thus detectable. Spectra of different backgrounds (conveyer belt and tile), OPP20/PE50 (transparent and printed in different colors) and the corresponding monolayer samples are presented in Fig. 2.

Fig. 2a shows that the transparent OPP20/PE50 sample has a spectrum different from background material (both conveyer belt and tile). Studies showed that spectra of PP and PE are characterized with peaks at 1190 – 1200 and around 1400 nm (Masoumi et al., 2012; Rani et al., 2019; Kumagai et al., 2002). From Fig. 2a is to see that the spectra of OPP20/PE50 on conveyer belt differ from spectra of the background with peaks at approx. 1160–1240 and 1300–1450 nm, which are typical characteristic peaks of PP and PE. The spectrum of OPP20/PE50 with tile as background has similar form but much higher intensities than directly on the conveyer belt. The reason is that the tile reflects more light comparing to black background, and the changes in intensity are thus higher. Since the used tile is NIR-inactive, changes in the intensity of the reflected radiation can be explained by the non-uniform irradiation from emitters or the increase in overall intensity, which lead to an amplification of already existing peaks that were undetectable at

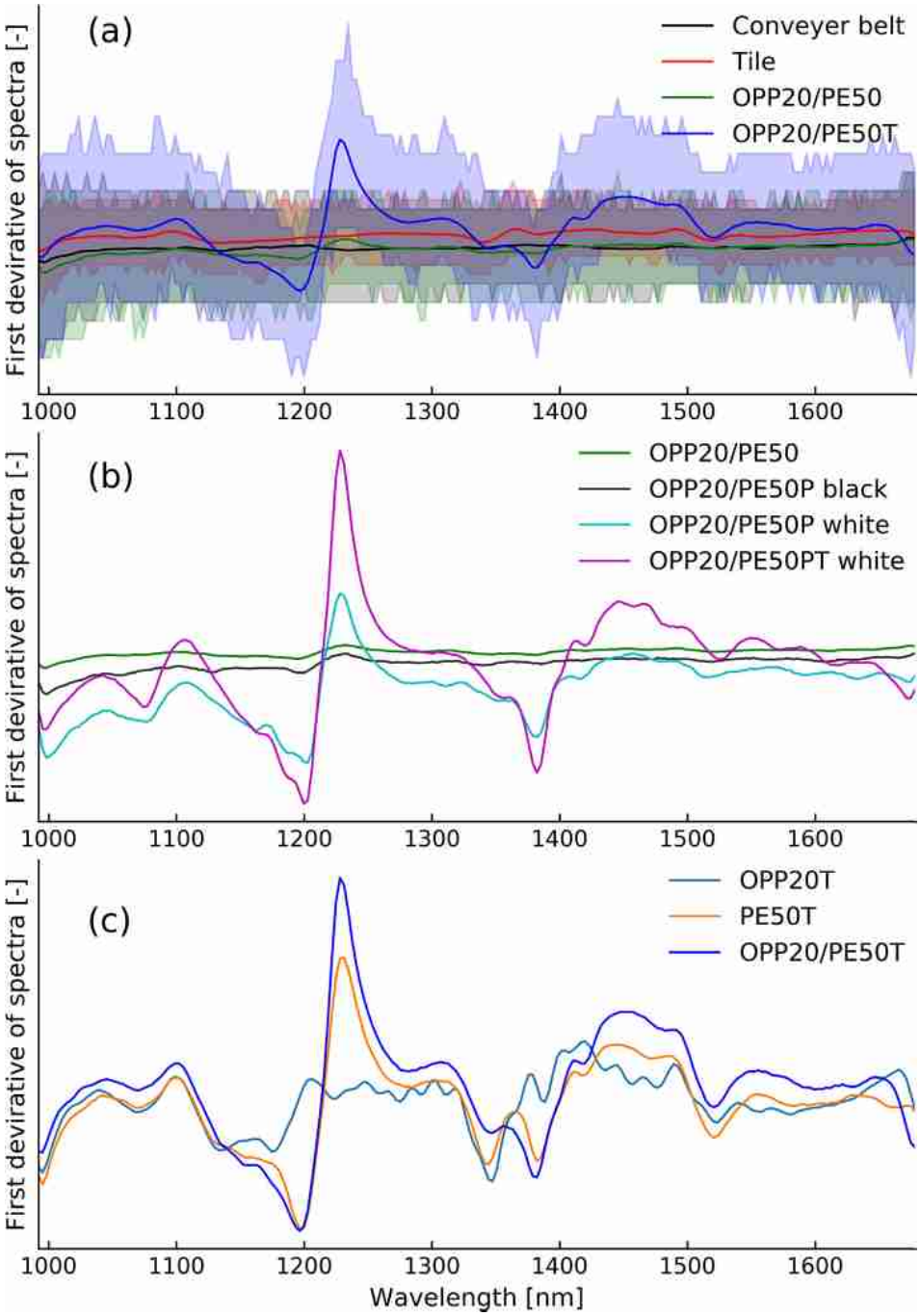

Fig. 2. Comparison of spectra. (a) Spectra of OPP20/PE50 and of different backgrounds (conveyer belt and tile) with variation (min. and max. values of 1600 pixels, colored regions); (b) spectra of OPP20/PE50 transparent and printed in different colors; (c) Spectra of OPP20/PE50 and the corresponding monolayer samples: OPP20 and PE50.

low reflection. Printed samples were also possible to be detected, see Fig. 2b. They have characteristic peaks at the same positions, and the printed color affects the intensity of the spectrum: samples printed in black have much lower intensities than samples printed in white. Like non-printed samples, samples on tiles have a much higher intensity than samples with only the conveyer belt as background. Analysis of other printed and transparent samples have shown similar results: the spectrum of samples printed in color (black excluded) has higher intensities than transparent samples, and pixels printed in bright colors have also higher intensities than dark colors. In addition, the spectrum of multilayer samples was compared to corresponding monolayer samples, see OPP20/PE50T with OPP20T and PE50T (Fig. 2c) as an example. The spectrum of OPP20/PE50T was similar to the spectrum of PE50T, but they differ at about 1350 nm. According to Kumagai et al., PP differs from PE with the peak at about 1350 nm (Kumagai et al., 2002), which means, the spectra of OPP20/PE50T have both characteristics of PP and PE. With differences in spectra, the samples (OPP20/PE50, OPP20, PE50 and background) were able to be classified to corresponding classes with a pixel-based classification accuracy of more than 95%. Misclassification happens mainly due to the noise and variation (cf. Fig. 2a) as well as areas which are printed in black.

In summary, all samples, except for some non-printed glossy PET12-containing samples, were able to be detected. They could be differentiated from background and monolayer materials on the basis of the characteristic spectra. Spectra of samples show much higher intensity when an NIR-inactive tile instead of a black conveyer belt is used as a background.

3.2. Qualitative analysis

Since samples were able to be detected and classified from background and corresponding monolayers, it is important to determine whether different material combinations can be identified and how different materials influence the spectra and the classification results. As a known material composition is necessary for the existing recycling processes for MPP, samples were classified according to different strategies, and the classification accuracies were evaluated to determine proper classification strategies for existing recycling processes.

3.2.1. Influence of plastic layers

Spectra of samples with different material combination were compared with each other and the samples were pixel-based classified. Fig. 3 shows the mean spectrum of different transparent PE50-containing samples (PE50, OPP30/PE50, PET36/PE50, OPA15/PE50) and EVOH-containing PE sample (PE/EVOH/PE90).

As illustrated in Fig. 3, PE-containing samples have similar spectra in most wavelength areas, but different material combinations show slightly different spectra. Each material has its own characteristic peak position, regardless of how thick the corresponding layer is. OPA/PE samples have a positive peak at about 1530–1550 nm, which assigned to the contribution from PA, as the most characteristic peak of PA locates at about 1460–1540 nm (Kumagai et al., 2002). For OPP/PE, the characteristic peak is located at about 1350 nm, which is the location of the spectral signature of PP, like mentioned in Section 3.1.2. There is a typical negative PET-peak at approx. 1650 nm for PET/PE, which corresponds to most characteristic peak of PET compared to other common plastics, e.g. PP, PE and PA (Masoumi et al., 2012; Huber et al., 2015; Rani et al., 2019). Even the EVOH layer, which is only 5 μm thick, shows a slight difference to PE which can be detected at about 1344 nm. This peak assigned to the contribution of EVOH, as EVOH is characterized with the peak at about 1300–1400 nm (Iwamoto et al., 2001). These slight differences were sufficient to classify them into their corresponding classes. The pixel-based classification accuracy with the used PLS algorithm for the material combinations mentioned above was 96.7% ± 0.5%. For samples without metalized layers, the layer sequence, i.e. which side is exposed to NIR sensor does not influence the spectrum and the classification results.

3.2.2. Metalized layer

The evaluation showed that a metallized layer has a specific effect on the spectrum. The samples OPP20/PETmet12/PE30 and OPP20/PETmet12/PP40 showed almost the same spectra, which means, the underlying layer does not influence the spectrum. Besides, depending on which layer is exposed to the NIR sensor, the spectra change according to the material of the layer above the metalized layer. Through comparison of spectra of PE30/PETmet12/OPP20, PE40/PETmet12/OPP20 and PE50/PETmet12/OPP20, it was determined that the plastic layers lying above the metallized layer show a more intensive influence on the spectrum with increasing thickness. If these layers are printed, this characteristic spectrum becomes more intensive and more similar to PE layers.

The analysis of samples with a metallized plastic layer showed that no radiation can pass through a metallized layer, which is also the purpose of application of metalized layer. Thus, no change in the spectrum occurs when different plastics with different thick-

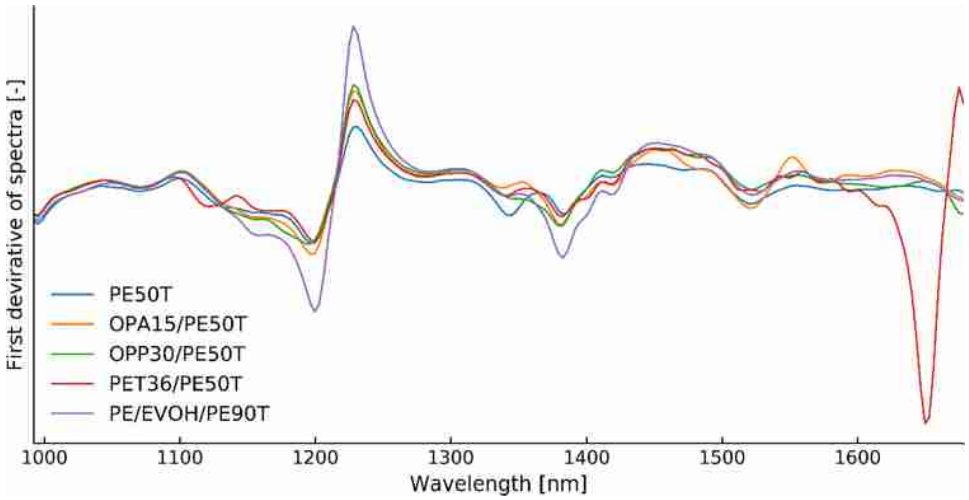

Fig. 3. Mean spectra of different PE50-containing samples and EVOH-containing PE sample.

nesses are placed under the metallized layer. For detection and classification, samples with a metalized layer are likely to be classified as materials above the metalized layer.

3.2.3. Classification according to material combination

All samples without two-layer PET12-containing multilayer (cf. Section 3.1.1) were classified according to the material combination. The samples were firstly pixel-based classified and the object-based classification accuracy was determined afterwards. Fig. 4 shows the classification result of non-printed transparent samples composed of two different materials (two-layer samples and EVOH-containing three-layer samples). For each sample, an area of 100*60 pixels is illustrated.

For classification of samples consisting of two different materials, the pixel-based classification accuracy was 96.3% ± 0.4%. Analysis and classification of all two- and three-layers samples without metalized layer showed that the classification accuracy of all samples was higher than 80%, which resulted in an object-classification accuracy for 24 non-printed two- and three-layers samples of 100%. In case all samples are classified to two-material combinations (the 8 classes in Fig. 4), the metalized-layer-containing and four-layer samples were classified to the class of their main composition, for example, PET12/PE/EVOH/PE90 was 78.4% classified as PE/EVOH/PE and 9.2% classified as PET/PE. For non-printed transparent samples, the pixel-based classification accuracy with tile as background is about 6% higher than with a black conveyer belt as background.

Printed samples can be classified with much higher accuracy when they are trained and classified without transparent samples. Their classification results were similar to transparent samples. However, as some area was printed in black, the pixel-based classification accuracy was 2–3% lower than transparent ones. In case non-printed and printed samples are analyzed together, the object-based classification accuracy with training all 120 samples was 67%. For this reason, printed and non-printed samples need to be separately analyzed. An integration of an RGB-camera for sensor-based MPP sorting is a possible solution to separate printed and non-printed samples.

The classification above showed that it is possible to classify MPP samples according to the main material combination. For different recycling processes, MPP can be separated with different sorting strategies. For the process of APK, samples can be classified to two classes: PA/PE and others. In this case, our best PLS model achieved an object-based classification accuracy of 100%, when non-printed and printed samples were analyzed separately. For CreaSolv, samples can be sorted according to PP/PE-containing samples and samples without PP and PE. In case samples are classified to PE-containing and not PE-containing instances, the object-based classification accuracy was 97.3%.

3.3. Quantitative analysis

With the thickness of one material increases, the volume proportion of this material increases. On the basis of the spectra of samples with the same materials but different proportions, the possibility for a quantitative analysis is determined. Through analysis of spectra of OPA15/PE30, OPA15/PE40, OPA15/PE50, OPA15/PE100 and PE100, it was determined that the higher the thickness of the whole sample, the higher is the intensity at specific peaks. With increasing percentage of PE, the spectra are getting more similar to the PE monolayer spectrum. For non-printed samples, our best regression model with the used PLS algorithm for spectrum and volume percentage of PE with a black conveyer belt as background achieved an R^2-score of 0.876. For OPP/PE samples, the R^2-score of the regression model of five samples was 0.893. In case tile is used as background, the R^2-score is approx. 3% higher for both material combinations, see Table 2.

For prediction, the results of OPA/PE samples show that with a given spectrum, the percentage of one material in a transparent sample with tile as background can be determined with a root mean squared error of 2.3 vol-% and a mean absolute error of 1.9 vol-%. The prediction for 6000 pixels of each sample is shown in Fig. 5. Besides, the R^2-score is approx. 2% higher with NIR-inactive material as background.

3.4. Detection of post-consumer MPP

3.4.1. Material presentation

Samples were folded to simulate bag packaging. For transparent samples, the higher the thickness, the higher the intensities and the easier is the classification. For transparent samples which were folded one time, the pixel-based classification accuracy has increased from 91.3% ± 0.7% to 93.7% ± 0.6% for two- and three-layers samples. With increasing folded times, the thickness of the sample increased, and the classification accuracy of the samples is getting higher. For printed samples, intensities almost did not change, and the characteristic peaks are always present. As a result, the classification accuracy changed little. Therefore, folded samples are still possible to be classified according to the material composition, and folding samples also contributes to a better classification for non-printed samples.

3.4.2. Post-consumer MPP

After mixing with LWP waste, samples got impurities on the surface, which resulted in less overexposed areas than clean sam-

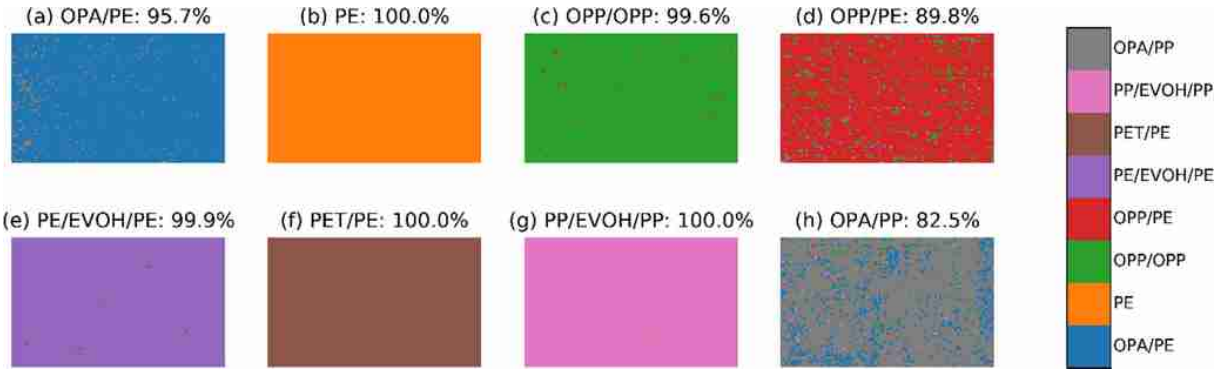

Fig. 4. Classification results of non-printed transparent OPA/PE, PE/PE, OPP/PP, OPP/PE, PE/EVOH/PE, PET/PE, PP/EVOH/PP, OPA/PP (top of each picture is the true class of this sample and the classification accuracy, each color represents a predicted material type).

Table 2
Regression model performance for OPA/PE and OPA/PP with different proportions.

Material combination	OPA/PE (conveyer belt)	OPA/PP (conveyer belt)	OPA/PE (tile)	OPA/PP (tile)
R^2-score	0.876 ± 0.016	0.893 ± 0.014	0.905 ± 0.005	0.918 ± 0.006
Root mean squared error [Vol.-%]	3.6% ± 0.2%	3.2% ± 0.2%	2.3% ± 0.1%	2.1% ± 0.1%
Mean absolute error [Vol.-%]	2.9% ± 0.2%	2.7% ± 0.1%	1.9% ±0.05%	1.8% ± 0.04%

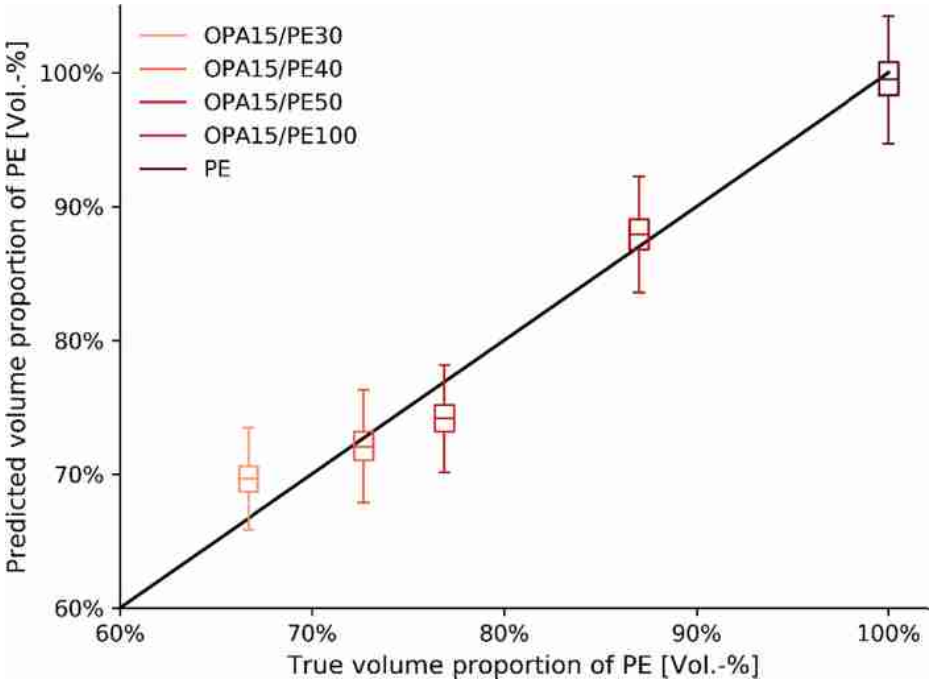

Fig. 5. Prediction of volume proportion of PE with regression model; Box: 25% and 75% quantile, Whisker: 0.5% and 99.5% quantile.

ples: the proportion of overexposed area was reduced from 9.3% in all clean samples to 3.7% in samples after mechanical pretreatment. Besides, the surfaces of low-thickness PET12-containing multilayer samples were contaminated and are thus, less glossy than clean samples. As a result, the spectra are no more wave-like and the pixels are possible to be analyzed. Fig. 6 shows the mean spectra of samples before and after mixing with LWP waste.

As illustrated in Fig. 6a, the spectra of samples after simulation of mechanical treatment show a strong difference compared to those of clean PET12/PE30 samples. For samples which were placed in water before mixing with LWP waste, the spectra have minor fluctuation (Fig. 6a), but also show typical characteristic peaks of PET/PE (peaks at about 1240 nm and 1680 nm). For samples which were placed in oil before mixing with LWP waste, the spectra show little variation and the same characteristic peaks. In both cases, the spectra could be classified to right classes because of the existence of characteristic peaks. Samples without PET12 layer after mechanical treatment simulation have similar spectra like clean samples and were still possible to be classified, see Fig. 6b. For classification according to material composition of two- and three-layers non-printed transparent samples after mechanical treatment simulation, pixel-based classification accuracy was 95.7% ± 0.4% and object-based classification has an accuracy of 100%.

For samples with a metalized layer, the spectra after mechanical treatment are almost identical to clean ones. A metallized layer, like before, does not allow any radiation to pass through. As a consequence, layers under a metalized layer are invisible for NIR-sensor and do not have any influence on the spectrum.

In summary, mechanical treatment simulation in drum sieve with LWP waste has an influence on the surface of samples. For glossy samples which have direct and non-dispersive reflection, the detection and classification benefits from the mechanical treatment, since the surfaces are less glossy afterwards. For other samples, mixing with LWP waste has almost no influence on the detection and classification. The simulation of food contact (in water or in oil) has little influence on the spectra of samples and their classification. Surprisingly, the collection and preprocessing process show a positive influence on the classification of MPP. On the basis of the classification results, it can be concluded that from a classification point of view, sorting of post-consumer MPP is achievable.

4. Conclusions

In this research, the possibility to classify and sort post-consumer MPP has been investigated. Our results show that from the classification point of view, sorting is possible. Firstly, clean samples were studied. Except for very smooth and glossy samples and overexposed pixels, clean samples were able to be detected and classified. Samples were then contaminated with water and oil, mixed with LWP waste in a drum sieve afterwards to simulate the mechanical treatment during collection and preprocessing at a sorting plant. Spectra and classification results illustrated that the samples were still possible to be detected and classified. For glossy samples, mechanical treatment even contributed to a better classification result. With different classification strategies, e.g. sorting

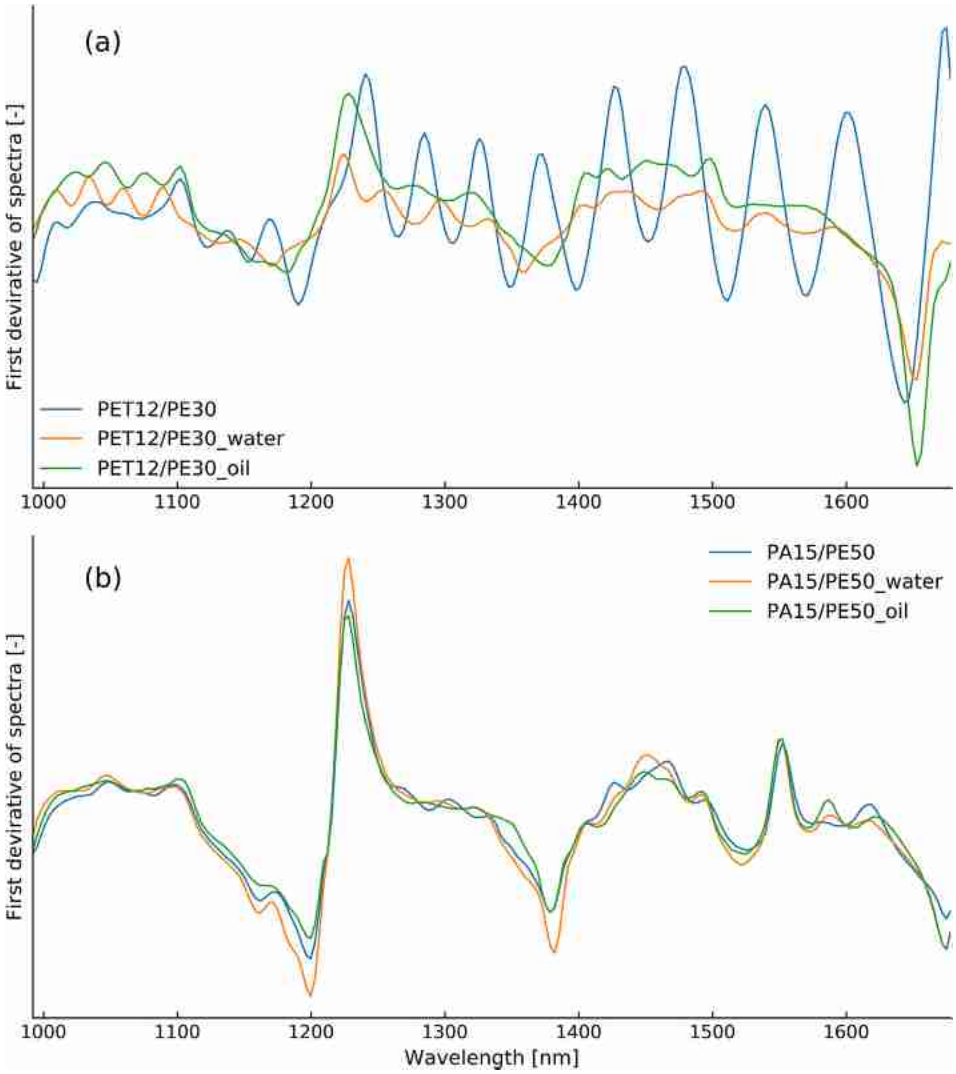

Fig 6. Mean spectra of samples before and after mechanical treatment simulation (in water/oil and with LWP waste); (a): PET12/PE30, (b): OPA15/PE50.

according to material components or to main composition, the object-based classification accuracy was higher than 92%. Depending on the recycling process, post-consumer MPP is possible to be classified according to the requirements. For both existing processes which are suitable for post-consumer MPP (Newcycling and CreaSolv), MPP can be sorted respectively to PA/PE and others as well as to PP/PE-containing samples and samples without PP and PE. In both cases, our best model has achieved an object-based accuracy of 96.2%. In addition, analysis results showed a possibility to determine the proportion of each material component. However, due to the limitation of samples, a more precise determination of accuracy needs to be investigated with more samples.

On the basis of the analysis and classification results, suggestions for a more recycling-friendly product design of MPP can be given. For classification, the less material types in a MPP, the higher is the classification accuracy and the easier is the classification to the detailed classes. Therefore, with ensuring the functionality of MPP, less material types should be used in one packaging. Moreover, as areas printed in black were more difficult to be classified than other bright colors, it should be avoided to print MPP in black if possible. To reduce non–detectable cases (e.g. wave-like spectra and overexposed areas), very glossy and highly reflective surfaces should be avoided. Furthermore, producers should reduce the use of metalized layers, since the underlying layers are not able to be detected.

As all the samples in this study are with the same additive layers, the influence of additive layers to NIR spectra was not determined. In further research, the influence of different additive layers can be investigated. Besides, the classification of MPP can be upscaled to sorting with industrial sensor-based sorters. More samples, especially real post-consumer MPP waste, can be used as input material to validate the feasibility of classifying waste films. For a better sorting result, information from RGB-sensors may be incorporated for the purpose to separately process printed and non-printed MPP, as the classification accuracy was much higher if printed and non-printed samples were individually classified. To achieve a higher classification accuracy for non-printed samples, an NIR-inactive background is helpful. Furthermore, the separation unit needs to be adjusted, as films have a different movement on conveyer belt and an unusual flight behavior. Since films have a much lower grammage comparing to three-dimensional plastics, the throughput of sorting MPP needs to be considered, and on the basis of this, the economical effort for the recycling of MPP needs to be evaluated.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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