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Influence of long-term natural degradation processes on near-infrared spectra and sorting of post-consumer plastics

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ABSTRACT

The large-amount production and application of plastics since the 1950s has led to different environmental problems, and the production amount is still increasing. In 2015, 79 wt% of all plastic waste was accumulated in landfills or the natural environment. Due to their negative influence to the environment, the problems of landfilling and marine litter need urgent treatments. Accordingly, measures like excavation of landfill sites and ocean clean-ups were conducted to reduce their environmental influences and move further towards a closed loop of material cycles. For a possible recycling, the valuable material fractions need to be separated from other materials. Besides, to ensure a high-quality recycling and enable the different recycling processes of plastics in different degradation levels, it is necessary to separate degraded and non-degraded plastics. In this study, the possibility to classify and sort landfill and marine litter plastics is investigated. For this purpose, waste plastics from different origins (lightweight packaging (LWP) waste, landfill, and marine litter) were collected and analyzed with the state-of-the-art technology in sorting plants: near-infrared spectroscopy. With self-developed programs, the classification possibility and performance was determined. The classification accuracy of degraded plastics (from landfill and marine litter) is improved from > 75% to > 97% through adjusting the sorting recipe. Besides, the long-term degraded plastics under natural environment were able to be separated from LWP waste: the same kind of materials can be classified according to their origin (LWP or after long-term degradation), which makes a quality control possible and enables an extra treatment for degraded plastics.

1. Introduction

Plastic is one of the most crucial and frequently used materials and is applied in almost all industries (Ashurst, 2016). In 2019, 368 million tons of plastics were produced worldwide and the production amount is still increasing (PlasticsEurope, 2020). The high amount plastic production and consumption lead to different environmental problems such as landfilling and ocean pollution. In 2015, about 6,300 million metric tons of plastic waste was generated, however, only 21 wt% was recycled or energetically recovered and the other 79 wt% was accumulated in landfills or the natural environment (Geyer et al., 2017). As a large-scale plastic production dates back to the 1950s (Geyer et al., 2017), the caused environmental problems need an urgent treatment. To achieve a sustainable development and intact environments for future generations, plastic waste streams need to be minimized and adequately treated with minimal environmental footprints by following suitable strategies of the circular economy through (i) avoiding unwanted plastic leakage, and (ii) enhancing material recovery by minimizing plastic landfilling and shifting towards closed material cycles wherever possible (Kirchherr et al., 2017; ten Brink et al., 2018).

Ocean clean-ups (Hee et al., 2021) and (enhanced) landfill mining (Jones et al., 2013; Vollprecht et al., 2021) are suitable measures to remove existing plastic pollution and avoid further plastic accumulation and damage to the environment. Ideally, recovered plastics from such measures could not only be made harmless but also reprocessed and further utilized to gain additional environmental benefits by reintroducing recovered plastics as secondary raw materials into the anthropogenic material cycle (Jones et al., 2013). Reprocessing of excavated waste and marine litter can be conducted with means of (i) mechanical recycling, (ii) chemical recycling, and (iii) energy recovery, depending on specific characteristics of the recovered plastics (Osterath, 2020).

According to life cycle analysis, mechanical recycling is the most

Abbreviations: BC, beverage cartons; LWP, lightweight packaging; NIR, near-infrared; PE, polyethylene; PET, polyethylene terephthalate; PLS, partial least squares; PP, polypropylene; PPC, paper and cardboard; PS, polystyrene; UV, ultraviolet radiation.

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efficient and environmental friendly treatment for plastic waste (Osterath, 2020). Studies have confirmed that mechanical recycling of excavated plastic waste is possible, if quality requirements (such as low degradation and limited contamination level of the considered plastics) are fulfilled (Canopoli et al., 2018). Furthermore, upcycling through pyrolysis is an alternative option for excavated plastics and marine litter, which requires a series of extra pretreatments (Breyer et al., 2017; Hee et al., 2021). In the case of a long-term degradation of plastics with high impurities, only energetic recovery is possible due to suboptimal material characteristics, high material contamination and degradation levels (Bosmans et al., 2013; Canopoli et al., 2018; Quaghebeur et al., 2013; Zhou et al., 2014).

Excavated landfill waste and waste from ocean clean-ups are heterogeneous mixes of different materials with often high impurity contents (Garcia Lopez et al., 2018). Thus, it is necessary to generate plastic preconcentrates that meet the aforementioned minimum quality requirements by sorting out impurities and separating different polymer types (Canopoli et al., 2018). As different plastic articles may have different retention times in the natural environments (i.e., different time intervals from landfilling/leakage until excavation/recovery) and may have been exposed to different degradation processes during those retention times, the degradation level of different plastics articles from similar recovery times may vary strongly. Besides, the radiation degradation induced by ultraviolet radiation (UV) (e.g., for marine litter) is not negligible (Celina, 2013), and the existence of residues, additives, or metals (e.g., for landfill plastics) may accelerate/initiate plastic degradation. Depending on the environment, different kinds of degradation (e.g., photodegradation, auto-oxidative degradation, thermooxidative degradation, thermal degradation, and biodegradation) occur: under specific conditions, polymers are broken down into shorter chains (mainly by microorganism enzymes) or new chemical bonds (e.g., ROOH) are formed (mainly initiated by UV and heat) (Canopoli et al., 2018). Therefore, separating plastic articles with lower degradation degrees (suitable for mechanical recycling) from plastic articles with higher degradation degrees (not suitable for mechanical recycling) is necessary.

1.1. Sensor-based plastic sorting

For commingled plastic waste, industrial-scale sorting of plastics by polymer type and impurity removal has been state-of-the-art for more than two decades (Feil and Pretz, 2020). This existing infrastructure could also be used for the pretreatment of excavated landfill waste (Maul et al., 2014) and plastic waste from ocean clean-ups. Modern sorting plants for post-consumer commingled plastic waste consist of several preconditioning steps (e.g., shredding, sieving, wind-shifting, magnet sorting and eddy current separation), followed by cascade of sensor-based sorters which separate different polymer types and remove impurities on the basis of material-specific near-infrared (NIR) spectra (Feil and Pretz, 2020). Modern sorting plants often contain more than 20 sensor-based sorters that separate commonly used plastics such as polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP), and polystyrene (PS). Sophisticated sorting recipes, task-specific classification algorithms and machine settings allow state-of-the-art sensor-based sorters to achieve high yields and purities of the generated preconcentrates (Gundupalli Paulraj et al., 2016).

Studies have confirmed that sorting of landfill material and marine litter with NIR spectroscopy is possible with specific self-developed sorting recipes trained with spectra of degraded plastics (Hee et al., 2021; Küppers et al., 2019a). However, the NIR-spectra of degraded samples may differ from that of non-degraded post-consumer plastics, which would reduce the sensor-based sorting performance and make it necessary to adapt existing sorting recipes used in state-of-the-art sorting plants. Furthermore, due to the short life-cycle times of (non-degraded) post-consumer plastics, separating plastics based on their degradation level is currently not applied in modern lightweight packaging (LWP) sorting plants. Therefore, it is necessary to research (i) how the NIR spectra change through natural degradation processes, (ii) how these changes influence the classification and sorting performance with existing sorting infrastructure for post-consumer plastics, and (iii) whether sensor-based differentiation between degraded and non-degraded plastics is possible.

1.2. Related work

Detection and classification of plastic aging/degradation under laboratory condition with NIR spectroscopy was investigated by several studies (Alassali et al., 2018, 2020; Chen et al., 2021). In these studies, the aging process of virgin plastics was conducted by thermally driven degradation process under laboratory conditions. It was determined that the aging of plastic samples was able to be detected through NIR spectral analysis (Alassali et al., 2018) and the predicted aging level of the samples had a good collinearity to aging time (Alassali et al., 2020). Chen et al. (2021) showed that the degradation of bioplastics under laboratory conditions was possible to be detected and a classification of degraded and non-degraded polylactic acid samples was achievable.

However, Celina (2013) indicated that the accelerated aging/degradation processes under laboratory conditions and natural ambient environmental conditions can be greatly different and show different degradation influence on plastic samples. Furthermore, the contamination on plastic surface during the natural degradation process could influence the detection, as surface conditions influence the NIR-based detection and classification (Küppers et al., 2019b). This difference was confirmed by Signoret et al. (2020a,b) through comparing the mid-infrared spectra of accelerated and natural photodegradation of plastics. Reviewing the aforementioned studies shows that there is a research gap of investigating the detectability of samples which were degraded under natural ambient environmental conditions with NIR spectroscopy.

1.3. Aim and scope

In this study, the possibility to detect the degradation of plastic waste under natural circumstances is investigated with NIR spectroscopy. For this purpose, samples which were collected from LWP sorting plant, landfill site and marine litter were analyzed. The classification algorithms were trained with valuable LWP materials which were collected from LWP sorting plants to simulate state-of-the-art sorting plant recipe for LWP sorting. Through pixel-based classification analysis, the possibility to sort landfill and marine litter plastic waste with currently applied sorting recipes in LWP sorting plants was determined. In addition, the possibility for separation of LWP waste and degraded plastics was investigated.

2. Materials and methods

2.1. Materials

To investigate different degradation levels, post-consumer LWP plastic waste with short lifetime, landfill plastics, and marine litter were analyzed. For an accurate comparison of the spectra, the valuable 3D material fractions for sorting plants (PET, PE, PP, and PS) from all three origins were analyzed.

Post-consumer plastic waste was collected from the product fractions of the lightweight packaging sorting plant Hündgen Entsorgungs GmbH & Co. KG (Swisttal, Germany). As more product fractions can be generated in sorting plants, all possible product fractions (beverage cartons (BC) and paper and cardboard (PPC) in addition to PET, PE, PP and PS) were used for training algorithms to avoid incorrect classification of samples to other production fractions. Detailed information of the LWP sampling and preprocessing steps can be found in (Kroell et al., 2021).

Landfill mining plastics were excavated from the landfill site at the

municipality of Mont-Saint-Guibert (Walloon Bravant, Belgium). The excavated waste was deposited between 1958 and 1985, i.e. the samples were degraded for more than 30 years. After excavation, the materials were fed into a ballistic separator to separate 2D and 3D plastics and then dried under laboratory conditions to exclude influence from water, as water may influence the NIR spectra (Küppers et al., 2019b). Detailed information of the landfill site and preprocessing steps of the landfill mining materials can be found in (Garcia Lopez et al., 2018).

Plastics from marine litter were collected from the islands Sylt and Norderney at the North Sea in Germany. The samples were washed up on the beach and randomly selected. Details to sampling site and collection process can be found in (Hee et al., 2021).

All the materials used in this study are in their original state, and plastics from landfill mining and marine litter were degraded under different conditions. The materials were manually sorted according to polymer type, and for each material type, 10 to 15 samples were randomly selected from each origin for further investigation.

2.2. Test rig

The capturing of NIR spectra was conducted with a Helios-G2-320 NIR sensor from EVK DI Kerschhaggl GmbH (Raaba, Austria). The spectral range was approx. 930 nm to 1700 nm with a spectral resolution of 3.1 nm/band. 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. With a belt width of 400 mm, the achieved spatial resolution is 1.25 mm/pixel.

2.3. Data analysis

The spectral data of all the samples was analyzed with self-developed analysis programs. For each kind of sample from different origins, the spectra data were extracted and saved in individual data frames. Depending on the classification cases, different data frames were used for training and classification.

2.3.1. Spectra extraction

For the training, classification and further analysis, spectral data of pixels from samples were extracted from background to exclude influences from background. Besides, saving all spectral data of the same material from the same origin (LWP, landfill, or marine litter) simplifies the determination of pixel-based classification accuracy.

As some samples are in irregular form and have impurities on the surface, an automatic data extraction of object pixels is not possible for all samples in the datasets. Transparent samples and impurities on the conveyor belt lead to an incorrect automatic detection of the objects. Therefore, data extraction of all samples was conducted with a semi-automatic process. For each sample, the first derivative of the reflection spectrum of each pixel was firstly calculated, as it better shows the change in reflectance intensities. A principle component analysis with a component number of 3 was then applied to the first derivative of pixels in this sample, through which the three most important principle components were extracted. These three components are weighted by 6/9, 2/9 and 1/9 respectively to emphasize the variance explanation by each principle component, and accordingly, an grayscale image (for a better illustration of the contour, the images in Fig. 2 are shown with color) of the sample is generated, as shown Fig. 2 left. For this image, an automatic edge detection was conducted and ideally, the objects should be detected from background (Fig. 2a). However, for samples in irregular shape or are transparent (see Fig. 2b as an example), the object detection does not work perfectly due to similar intensities. Depending on the object detection results, the user can choose whether a manual selection of area of interest is needed. For this purpose, the sample is shown with detected edges in blue lines, see Fig. 2 left. In the case that the edge detection works (Fig. 2a left), a binary erosion was applied to the detected objects to avoid edge pixels. In the case that a manual selection is necessary, an area (area with red line as contour) could be selected manually by mouse clicks. The overlap of the area from edge detection and manually selected area is the area of interest. Like without manual selection, a binary erosion was applied to the overlap. In both cases, the spectral data of pixels in the area of interest (yellow area in Fig. 2 right) were extracted and saved.

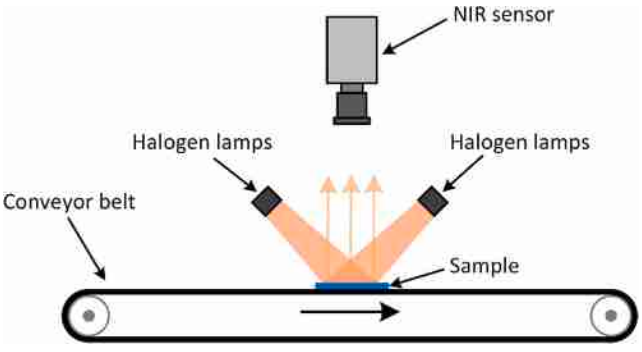

Fig. 1. NIR sensor and illumination setup.

The extracted spectral data from the same materials and the same origin were saved as individual data frames and labeled with material type, origin and object number. An overview of instance numbers in each data set is shown in Table 1. The reason for less pixels for marine litter was its smaller object size compared to other two origins.

2.3.2. Spectra classification

For the analysis, the first derivative of the spectra data was used to enhance the visual resolution and to correct the baseline. Partial least square (PLS) algorithm from scikit-learn v0.24.1 (Pedregosa et al., 2011) was used for classification, and different component number was used. For training PLS algorithms, 1500 pixels each class were randomly selected from corresponding materials. For example, to determine the classification possibility of degraded samples with currently applied sorting recipe in LWP sorting plants, in total 9000 pixels (1500 pixels each for PET, PE, PP, PS, BC and PPC) were selected for training process and were divided into 70% training and 30% test data. The classification possibility and performance were estimated based on the pixel-based classification accuracy.

3. Results and discussion

The spectra were analyzed firstly to investigate whether there is difference between non-degraded samples and samples after long-term degradation under natural environment. The samples were then pixel-based classified with current recipe from a sorting plant, and it was investigated whether it is possible to classify samples with and without degradation.

3.1. Classification of degraded samples with LWP sorting plant recipe

The spectra of same material type from different origins were analyzed firstly. Fig. 3 shows the spectra of PE from different origins as an example. From each data frame of different origin, 300 pixels were randomly selected, and their mean spectrum and variation of all 300 pixels are shown.

Generally, the position of most characteristic peaks did not change, and the form of the mean spectra are similar to each other, for example, the characteristic positive peak of PE at about 1240 nm and peaks at about 1150 nm–1220 nm. However, the level of different peaks shows difference and some characteristic peaks become weaker or even disappeared. As shown in Fig. 3, the peak of PE from LWP at about 1360 nm was much weaker for PE in landfill and disappeared for PE in marine litter. The analysis of other material types showed similar results: most

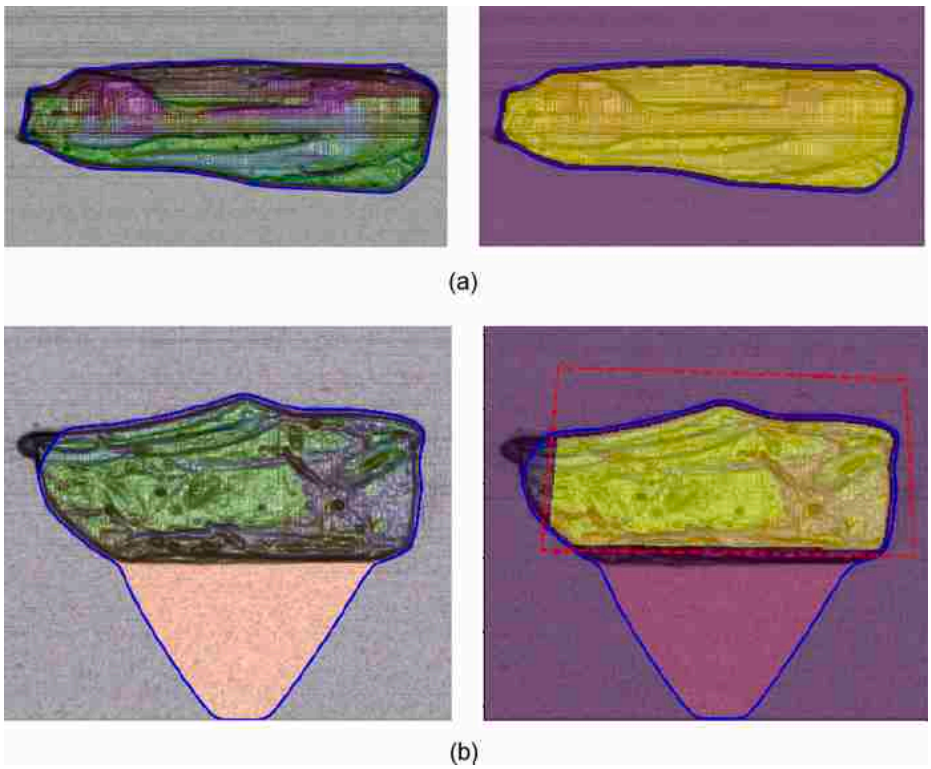

Fig. 2. Spectra extraction with self-developed semi-automated process. (a) Data extraction without manual selection; left: automatic object detection result; right: extracted area after erosion (in yellow). (b) Data extraction with manual selection; left: automatic object detection result; right: extracted area (in yellow), blue line is the automatic detected contour and red line is manual selection.

Table 1
Number of extracted pixels from each origin.

	PET	PE	PP	PS	BC	PPC
LWP	249,212	312,987	211,057	111,514	19,400	18,702
Landfill	212,021	290,412	145,545	118,688	–	–
Marine litter	68,000	35,350	18,032	16,802	–	–

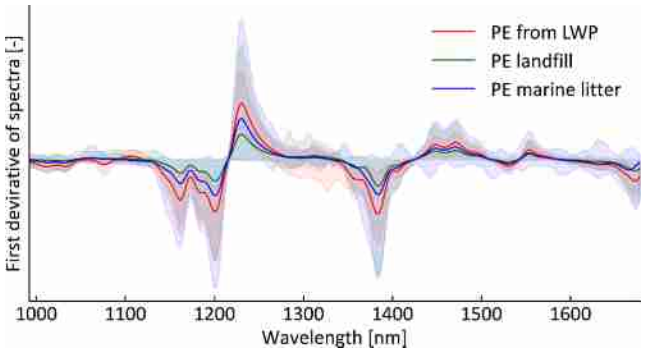

Fig. 3. Spectra of PE from different origins. Lines: mean spectrum of all 300 pixels; Colored range: spectra variation range.

characteristic peaks kept the same, but the peak intensity has changed. For PP, there is little difference in peak position and all the characteristic peaks existed but with different intensities. For PS, characteristic peaks in the range of 1350 nm–1420 nm disappeared in spectra of both landfill and marine litter samples. Besides, marine litter samples showed stronger difference to LWP samples in spectra than landfill samples despite the existence of impurities (there were more impurities on the surface of landfill samples than marine litter samples). The reason of less intensity of degraded samples could be the discoloration due to degradation or impurities on the surface, and the disappearance of characteristic peaks could result from the formation of new functional groups. For marine litter, the illustration of UV light may cause the more significant difference in spectra compared to landfill samples.

The samples from landfill and marine litter were classified with the algorithm trained with samples from the sorting plant. Due to the great difference in spectral data of different materials (PET, PE, PP, PS, BC and PPC), the accuracy on training data of a PLS algorithm with component number of 15 has reached higher than 99.96%. This algorithm was used for classifying degraded samples, and the classification accuracy of all pixels in each material class from landfill and marine litter is shown in Table 2.

Table 2 shows that the pixel-based classification accuracy corresponds to the results of the spectral analysis: great difference in spectrum led to relatively lower classification accuracy and the classification worked well for samples with slight difference in spectra. For example, the classification accuracy of PE from marine litter is much lower than that of PE from landfill, which may result from the disappearance of the characteristic peaks at about 1350 nm. The classification accuracy of PP is much higher due to little difference in spectra. Besides, the overall classification accuracy of marine litter samples was generally lower than that of landfill samples.

With current sorting recipe, the pixel-based classification accuracy of PE and PS from marine litter was relatively low. A more accurate

Table 2
Pixel-based classification accuracy of samples from different origin with current sorting recipe and a PLS component number of 15.

Origin	PET	PE	PP	PS
LWP	0.998	0.995	0.990	0.997
Landfill	0.947	0.948	0.995	0.898
Marine litter	0.901	0.772	0.972	0.755

classification and sorting of degraded samples can be achieved by training more spectral data from different origins. In the case that the training data are randomly selected from the three different origins, a pixel-based classification accuracy of higher than 97.2% can be achieved.

3.2. Classification of non– and degraded samples

To determine the possibility to detect whether the samples are degraded (from landfill or marine litter), algorithms were trained with samples in the same material but from different origins. For each material type, five classification algorithms were trained and applied. The first algorithms were used to determine the possibility to classify the same material from LWP and from landfill, for example, PE was classified to PE from LWP and PE from landfill. The other four algorithms were trained to classify samples into: LWP and marine litter; LWP and degraded samples (landfill + marine litter); LWP, landfill and marine litter; and marine litter and landfill. The pixel-based classification accuracy of each material class is shown in Fig. 4.

Although there is little difference in mean spectra of some materials from different origins (for example PP), the pixel-based classification accuracy of > 90% in classifying samples from LWP and degraded samples show that a classification of non-degraded and under natural environment degraded samples is possible. As both samples from landfill and marine litter were degraded, a classification between the two classes (from landfill and marine litter) was much more difficult with a classification accuracy of < 70%, and the classification accuracy of PP was < 60%. With the increase of the component number for PLS algorithms, the accuracy became slightly (about 5%) higher but is still insufficient for an accurate classification and sorting of all kinds of valuable materials.

From the classification point of view, a quality control of product quality is possible, as the degraded samples could be classified from non-degraded samples. However, the sorting of landfill and marine litter is not possible due to the low classification accuracy.

4. Conclusion

This study investigated the sorting possibility of long-term degraded plastics under natural environment: landfill and marine litter with NIR spectroscopy. Through spectral analysis, it was determined that the degraded samples have most of the characteristic peaks of the non-degraded samples, but show slight difference in position and levels of specific peaks. Different materials show also difference in spectra to various extend. The classification of landfill and marine litter plastics with current sorting recipe (algorithms trained with LWP materials) achieved a pixel-based classification accuracy of > 75%. Through adding spectra of degraded samples to training data, a classification with sufficient high accuracy (> 97.2%) is possible. Besides, a classification between degraded (landfill and material litter) and non-degraded samples is also achievable with a pixel-based classification accuracy of > 90%. The presented results correspond also to the conclusions from other studies that degraded samples are able to be detected with NIR spectroscopy and confirmed the degradation of landfill and marine litter plastics.

In practical applications, large scale sorting of landfill materials and marine litter in current sorting plant is, from a classification point of view, in principle possible. As degraded samples can be separated from the non-degraded LWP materials with high classification accuracy, different further processing steps for LWP and degraded plastic product fraction are theoretically realizable and thus, sorting landfill material and marine litter will most likely bring little influence to the sorting plant. In further studies, a large scale sorting of landfill and marine litter material flows should be conducted in sorting plants for upscaling of the presented results, and the quality of production fraction should be determined through large-scale sorting trials.

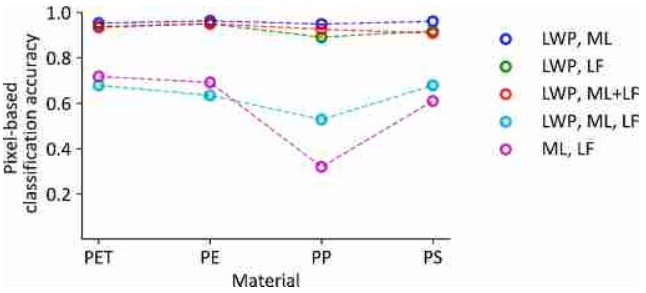

Fig. 4. Test score and classification accuracy of different materials according to degradation level with a component number of 15; ML: samples from marine litter; LF: landfill; Legend means classification between different classes, e.g. "LWP, ML + LF" means marine litter and landfill samples were grouped to one class and the classification was between LWP and the group of marine litter and landfill.

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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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