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RECYCLING-ORIENTED CHARACTERIZATION OF PET WASTE STREAM BY SWIR HYPERSPECTRAL IMAGING AND VARIABLE SELECTION METHODS

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ABSTRACT
The proposed study was carried out to develop a fast and efficient strategy for plastic waste sensor-based sorting in recycling plants, based on hyperspectral imaging (HSI), combined with variable selection methods, to produce a high-quality recycled polyethylene terephthalate (PET) flakes stream. Variable selection techniques were applied in order to identify a limited number of spectral bands useful to recognize the presence of other plastic materials, considered as contaminant, inside a stream of recycled PET flakes, reducing processing time as requested by sorting online applications. Post-consumer plastic samples were acquired by HSI working in the short-wave infrared (SWIR) range (1000 - 2500 nm). As a first step, the hypercubes were processed applying chemometric logics to build a partial least squares discriminant analysis (PLS-DA) classification model using the full investigated spectral range, able to identify PET and contaminant classes. As a second step, two different variable selection methods were then applied, i.e., interval PLS-DA (i-PLSDA) and variable importance in projection (VIP) scores, in order to identify a limited number of spectral bands useful to recognize the two classes and to evaluate the best method, showing efficiency values close to those obtained by the full spectrum model. The best result was achieved by the VIP score method with an average efficiency value of 0.98. The obtained results suggested that the variables selection method can represent a powerful approach for the sensor-based sorting online, decreasing the amount of data to be processed and thus enabling faster recognition compared to the full spectrum model.

1. INTRODUCTION

Plastic is one of the most used materials in daily life, thanks to its characteristics and versatility. Moreover, plastic waste is among the most diverse materials, making their recycling very complex (Ragaert et al., 2017). Due to the growing use of plastic, the amount of produced waste tends to increase over time, reaching an unsustainable pace for environmental reasons. It is thus necessary to develop and implement the best recycling strategies for plastic waste, guaranteeing high quality standards of the produced secondary raw materials and improving competitiveness with virgin polymers (Eriksen et al., 2018). Indeed, the quality of secondary raw materials resulting from the post-consumer plastic recycling process is highly dependent on the sorting efficiency throughout the plant line (Küppers, et.al, 2019). The purity degree of secondary plastics is certainly one of the most important quality characteristics required by the market (Faraca and Astrup, 2019). Traces of contaminant inside the recycled stream of a single polymer, both as other materials and other types of polymers, can affect the final properties of the secondary raw material. As a consequence, the identification and separation steps in mechanical recycling plants for homogeneous plastic production are critical (Alsewailem and Alrefaie, 2018). Accurate separation methods are needed for plastic recycling, which allow to minimize contaminant in recycled products (Serranti et al., 2011; Wu et al., 2013; Cucuzza et al., 2021). Hyperspectral imaging (HSI), coupled with chemometric logics, can represent an important tool to perform waste plastics identification and separation, such as polyethylene and polylactic acid (PLA) (Ulrici et al., 2013), polyolefins from building and construction

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waste (Serranti et al., 2012), PE and polypropylene (PP) from household waste (Serranti et al., 2015a), low density polyethylene (LDPE) and high density polyethylene (HDPE) recognition along with PP, polyvinyl chloride (PVC) and polystyrene (PS) (Bonifazi et al., 2018a), plastic containing brominated flame retardants (Bonifazi et al., 2020; Bonifazi et al., 2021a). HSI is a technology that integrates conventional imaging and spectroscopy, being able to attain both spatial and spectral information from an object (Gowen et al., 2007). HSI can be used in recycling plant as sensor-based sorting method. The potential application of HSI is widely demonstrated in the literature in different sectors, in addition to plastic waste, such as food control (Serranti et al., 2013; Bonifazi et al., 2021b), demolition waste (Serranti et al., 2015b; Bonifazi et al., 2018b; Trotta et al., 2021), hazardous materials (Bonifazi et al., 2018c; 2019). Therefore, HSI techniques represent an attractive solution for characterization, classification, quality control and sorting-online application also for polyethylene terephthalate (PET) waste streams.

The proposed study was carried out to build a fast and efficient strategy to produce a high-quality recycled PET stream, based on HSI working in the SWIR range (1000-2500 nm), recognizing PET flakes and other polymers (considered as a single class of contaminant). Indeed, it is essential to continuously refine research on PET recycling, as it is one of the most used polymers for food and beverage packaging, thanks to its physical and chemical characteristics (Welle, 2011). Furthermore, the use of HSI in the SWIR range ensures the recognition of the slight spectral differences between polymers, reducing errors of misclassification (Singh et al., 2017; Lorenzo-Navarro et al., 2021). In order to increase the data processing speed, as requested by the application of the classification logic to be utilized at industrial plant level, a variable selection approach was tested, allowing to select the most useful wavelengths for the identification of PET and contaminant inside the full investigated spectrum. In fact, in the sensor-based sorting process based on HSI the wavelength selection is strictly necessary in order to obtain an identification of materials with minor time and production costs. Different variable selection methods can be applied to near infrared data analysis (Yun et al., 2019; Mehmood et al., 2012). In the proposed case study, among the most used variable selection methods, i-PLSDA (Interval Partial Least Square Discriminant Analysis) and VIP (Variable Importance in Projection) were tested. In detail, a classification model based on PLS-DA in full spectrum mode was set up to identify classes of polymers, i.e., PET and other polymers considered as a single class of contaminants. Subsequently, i-PLSDA and VIP methods were applied. Finally, the results obtained were compared in order to identify the best variables selection method with the best predictive performance close to full spectrum classification model.

2. MATERIALS AND METHODS

2.1 The investigated samples

The plastic waste flakes used for this study, collected from a recycling plant, were randomly sampled and are representative of an online sorting scenario. They have an average size of 16 mm. In detail, the samples are constituted of PET flakes contaminated by small quantities of other polymers (such as PE, PP and PS) (Figure 1).

The dataset used to build and validate the model was composed by 55 PET and 55 contaminant flakes (Figure 1a), divided into a calibration (33 PET and 33 contaminant flakes) and a validation (22 PET and 22 contaminant flakes) set, as shown in Figure 1b. Finally, the classification model was applied to 3 different test sets (Figure 1c, d and e) composed by 389 randomly sampled plastic particles collected from the same output flakes stream of the calibration and validation datasets.

2.2 Data acquisition

Data acquisition was performed at the Raw Materials Laboratory (RawMaLab) of the Department of Chemical Engineering, Materials & Environment of Sapienza University of Rome, using the hyperspectral system SisuCHEMA XL (Specim, Spectral Imaging Ltd, Oulu, Finland), working in the SWIR region (1000 - 2500 nm), with the Imspector N25E spectrograph, spectral sampling/pixel: 6.3 nm; spectral resolution: 10 nm (30 μm slit), spatial resolution: root-mean-square spot radius <15 μm (320), field of view of 20 cm with 15mm lens, scanning speed (mm/s): 72.50 and active pixels: 320 (spatial) × 240 (spectral). The lighting was reproduced by applying a diffuse line illumination unit. Images were acquired performing a line by line scan of each investigated dataset. Instrument was equipped with an integrate hardware and software spectral calibration architecture. Image data were automatically calibrated by measuring an internal standard reference target before each dataset scan.

2.3 Hyperspectral data analysis

The acquired hyperspectral images were analyzed through the PLS_toolbox (ver. 8.8 Eigenvector Research, Inc.) running in the Matlab environment (version R2020a, The Mathworks, Inc.).

As this paper aimed to identify plastic contaminants inside a flow stream of PET, independently from their nature, a two-classes model was built, based on a PET class and a single class of contaminants. A data preprocessing to improve the collected spectral characteristics and an exploratory analysis of the data based on Principal Component Analysis (PCA), were performed. PCA was chosen to perform exploratory analysis about classes variability, to identify and remove outliers (Bro and Smilde, 2014). Partial least square discriminant analysis (PLS-DA) models in full spectrum and variable selection mode were defined and applied. PLS-DA is a supervised technique that needs a prior knowledge of the data (Barker and Rayens, 2003), which is based on the reduction of dimensionality through partial least squares regression (PLS-R) with discriminating characteristics (Ballabio and Todeschini, 2009; Ballabio and Consonni, 2013).

Different pre-processing techniques and combinations of algorithms were tested following the examples usually adopted in literature (Amigo et al., 2008; Rinnan et al., 2009; Amigo, 2010; Martens and Næs, 2011; Vidal and

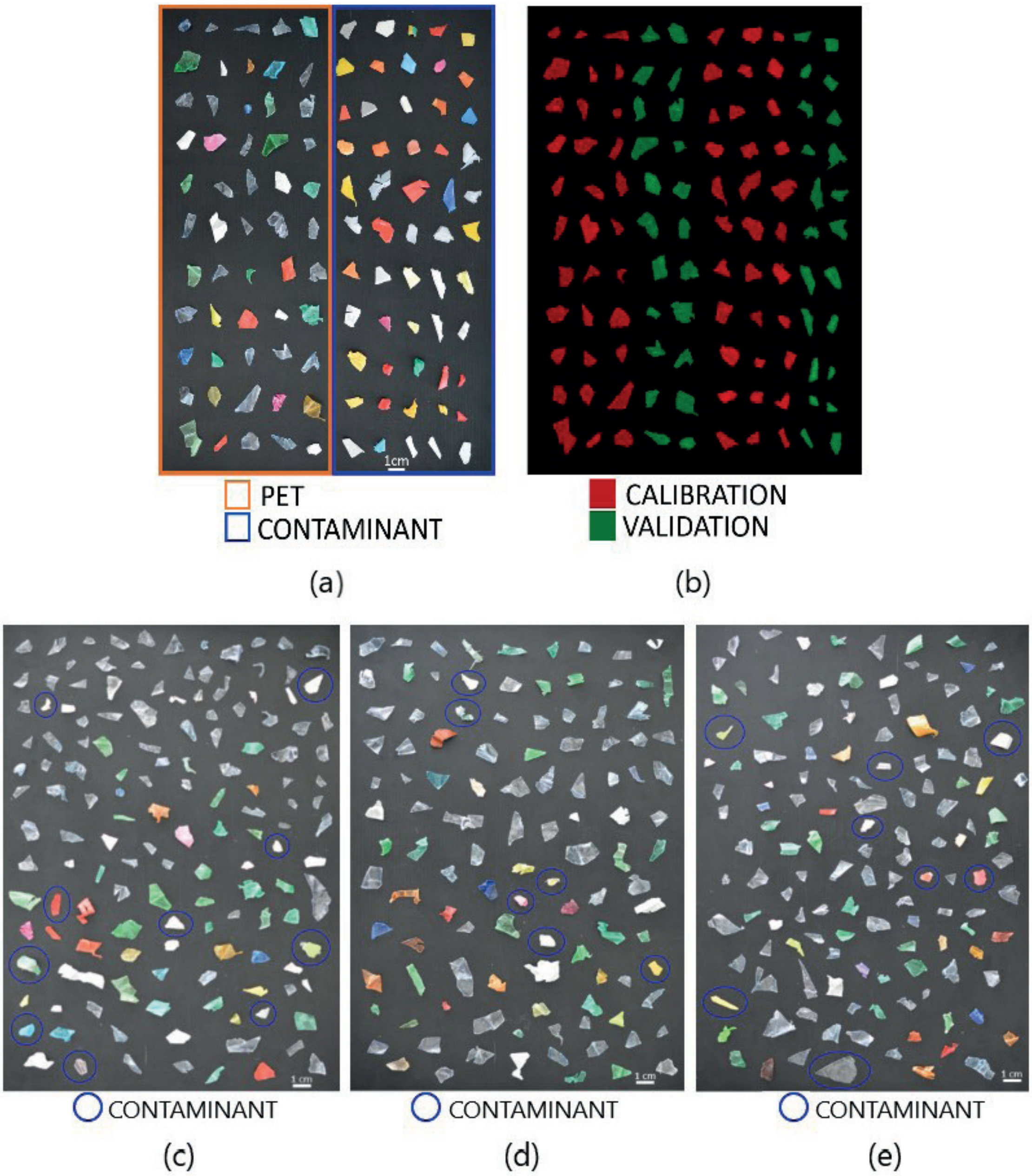

FIGURE 1: Source image of the investigated plastic samples divided in PET and contaminant flakes (a), false color image of the samples divided into calibration (red) and validation dataset (green) (b), source images of the randomly selected plastic flakes (contaminant marked by blue circles) for test 1 (c), test 2 (d) and test 3 (e).

Amigo, 2012; Calvini et al., 2016). Multiplicative Scatter Correction (MSC - median) method was used to remove scaling and offset effects, Savitzky - Golay smoothing (window 15 points) was chosen to delete high-frequency noise from samples, 1st derivative (polynomial order: 2, derivative order: 1 and window points: 21) was used to emphasize the characteristics of the bands and mean centering (MC) was applied to remove mean value and further improve the spectral differences between samples. The Contiguous Block (with a number of data splits equal to 10) cross-validation method was chosen (Figure 2) (Ballabio and Consonni 2013), in order to evaluate the complexity of models and to select the appropriate number of latent variables (LVs). Moreover, the optimal number of 3 LVs was decided by the smaller difference between RMSEC and RMSECV (Balage et al., 2018; Currà et al., 2019; Suhandy and Yulia, 2019).

Three PLS-DA classification models were built. The first model was developed using the full SWIR spectrum (1000-2500 nm), whereas the other two models using only wavelengths selected by i-PLSDA and VIP scores method. In detail, i-PLSDA selects a subset of variables by performing a sequential and exhaustive search for the best variable or combination of variables (Nergaard et al., 2000). The variable selection was made in "forward" mode, where the intervals was then included in the search, specifically

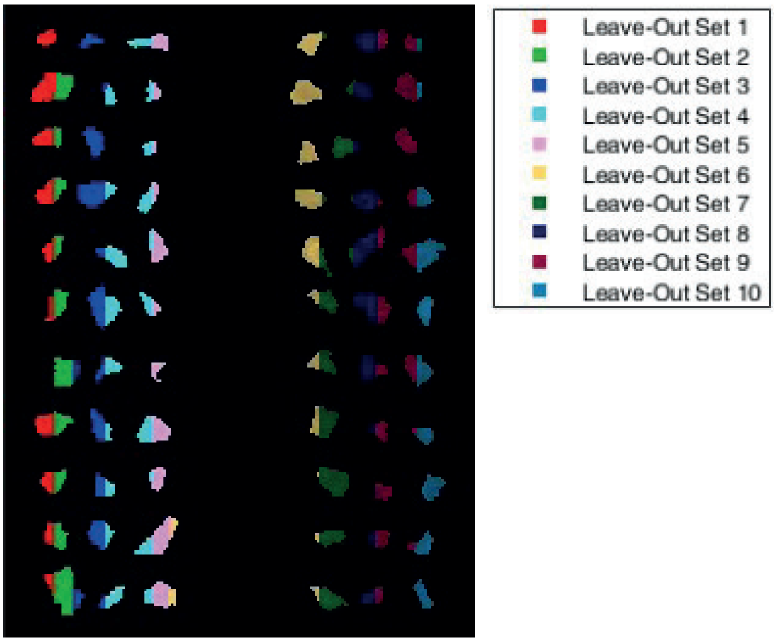

FIGURE 2: Results of the application of the contiguous block algorithm as cross-validation method on calibration dataset.

3 intervals with an interval size of 10. VIP scores estimate the importance of each variable in the projection used in a model (Chong and Jun 2005). VIP scores method was made considering 3 intervals with a dimension of 10 wavelengths. Such dimension was used as it corresponds to a wide spectral range of 60 nm, similar to that of common infrared broadband filters. In addition, 3 intervals were used to achieve an efficiency close to the full spectrum with a minimum number of wavelengths.

In order to compare the classification results, based on pixel detection, i.e., the attribution to one of the two classes (PET or contaminant) with reference to the number of pixels, the values of sensitivity, specificity and efficiency were calculated (equations 1, 2 and 3).

$$Sensitivity = \frac{True\ Positive}{(True\ Positive + False\ Negative)} \quad (1)$$

$$Specificity = \frac{True\ Negative}{(True\ Negative + False\ Positive)} \quad (2)$$

$$Efficiency = \sqrt{(Sensitivity \times Specificity)} \quad (3)$$

3. RESULTS AND DISCUSSION

3.1 Mean reflectance spectra

The average raw and pre-processed reflectance spectra of PET and contaminant classes are shown in Figure 3. PET spectrum was characterized by absorption bands of C-H_2 and C-H of the third harmonic region (1138 and 1180 nm), C-H of the second harmonic region (1400, 1660, 1720, 1830, 1910 and 1955 nm) and C-H stretching vibrations + C-H deformation of first combination region (2100, 2136, 2160, 2186 and 2261 nm). The mean spectrum of contaminant showed a complex fingerprint due the presence of different types of polymers. The main absorption bands detected for contaminant average spectrum were located around 1220, 1400, 1735 and 2320 nm. Finally, the pre-processed spectra allowed a differentiation between the two classes of materials.

3.2 Principal component analysis

PCA results are shown in Figure 4. Most of the variance was captured by the first two PCs, as shown in the PC1-PC2 score plot (Figure 4a), where PC1 and PC2 explained 74.61% and 13.76% of the variance, respectively. As shown in the PCA score plot, a separation was achieved between the clouds of PET and contaminants. In more detail, contaminant class showed higher variability, due to the presence of different type of polymers, being clustered across all quadrants in various groups mainly characterized by PC1 negative values. PET is characterized by a vertical cluster characterized by PC1 positive values. The loadings plot of PC1 and PC2 was shown in Figure 4b. The main PC1 variability is given by the wavelengths around 1170, 1375, 1705 and 2235 nm for positive values, while the negative values of PC1 are given mostly by the wavelengths 1265, 1475, 1865 and 2090 nm. PC2 is principally influenced by wavelengths around 1100, 1350, 1650 and 2100 nm for positive values, whereas negative values are more marked by wavelengths about 1235, 1470, 1750 and 2270 nm.

3.3 Full spectrum PLS-DA classification model

The results of the full spectrum PLS-DA classification model applied to the validation (Figure 5a) and test datasets (Figure 5b, c and d) are reported through the prediction images called “class predicted member”. PET and contaminant classes were correctly predicted in all datasets (cf. Figure 1 and 5).

The model correctly identified in the validation dataset 22 PET and 22 contaminant flakes (cf. Figure 1a, b and 5a), in the test 1 133 PET and 10 contaminant flakes (cf. Figure 1c and 5b), in the test 2 106 PET and 6 contaminant flakes (cf. Figure 1d and 5c), and in the test 3 126 PET and 8 con-

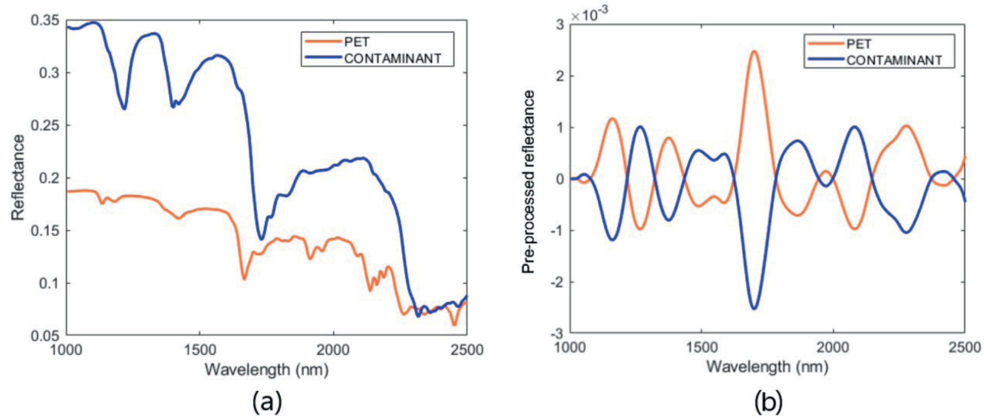

FIGURE 3: Average raw (a) and pre-processed reflectance spectra (b) of PET and contaminant classes.

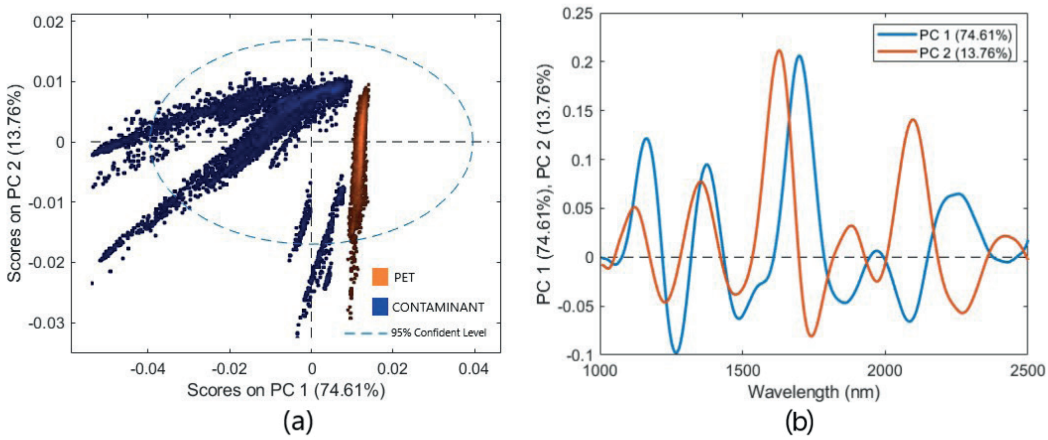

FIGURE 4: PCA score plot (PC1 - PC2) (a) and PCs loadings plot (b) of PET and contaminant classes.

taminant flakes (cf. Figure 1e and 5d).

The mean values of sensitivity and specificity, based on pixel detection, in calibration, cross-validation and prediction (Table 1) of validation, test 1, 2 and 3 datasets, showed the good performance of the model, with values of 1.00 for both classes.

Based on the application of i-PLSDA, the selected wavelengths were 1698-1754 nm corresponding to C-H of the second harmonic region, 1949-2005 nm and 2199-2255 nm corresponding to C-H stretching vibrations + C-H deformation of combination region of PET spectrum. Meanwhile the spectral bands selected using VIP scores method were 1138-1194 nm coinciding with $C\text{-}H_2$ and C-H of the third harmonic region, 1673-1729 nm corresponding to C-H of the second harmonic region and 2267-2324 nm correlated to C-H stretching vibrations + C-H deformation of combination region of PET spectrum. The spectral bands selected by i-PLSDA and VIP scores superimposed on the average spectra of the PET and contaminant classes are shown in Figure 6.

i-PLSDA classification model:. The results of i-PLSDA, in terms of prediction images, for the validation and test

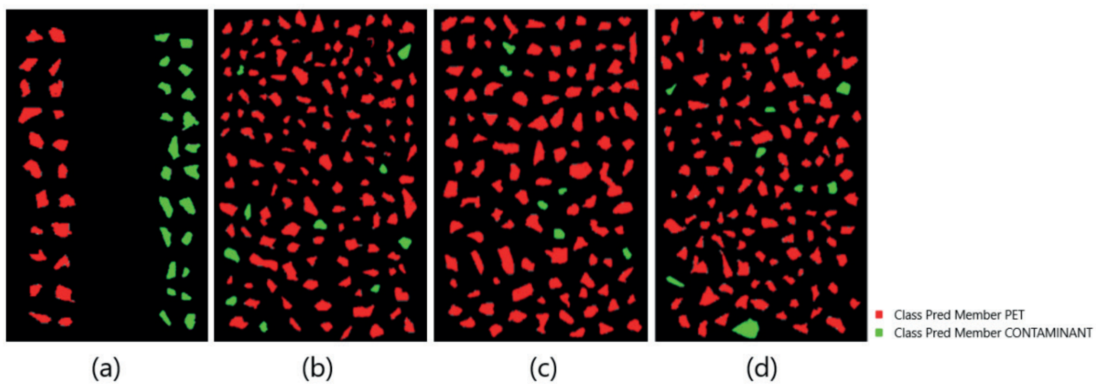

FIGURE 5: Full spectrum PLS-DA prediction maps for the validation (a), test 1 (b), test 2 (c) and test 3 (d) datasets.

TABLE 1: Mean performance values of full spectrum PLSDA classification for PET class in calibration, cross-validation and prediction phases. Selected LVs: 3.

	Sensitivity	Specificity
Calibration	1.00	1.00
Cross-validation	1.00	1.00
Prediction	1.00	1.00

datasets are shown in Figure 7.

PET and contaminant classes were correctly predicted, mainly in the validation dataset (cf. Figure 1a, 1b and 7a) and test 3 (cf. Figure 1e and 7d), except for some pixels due to border-effect (Figure 7b, 7c and 7d) and few samples attributed to the erroneous class (marked by yellow circles in Figure 7b and 7c), i.e., in the test 1: 4 badly assigned samples (cf. Figure 1c and 7b) and in the test 2: 2 badly assigned samples (cf. Figure 1d and 7c). The classification performances, based on pixel detection, obtained by i-PLSDA, shown in Table 2, revealed ranging values of sensitivity and specificity in calibration, cross-validation and prediction from 0.90 to 0.98.

VIP scores: The classification results obtained by the application VIP are shown in the prediction maps (Figure 8). PET and contaminant classes were correctly predicted,

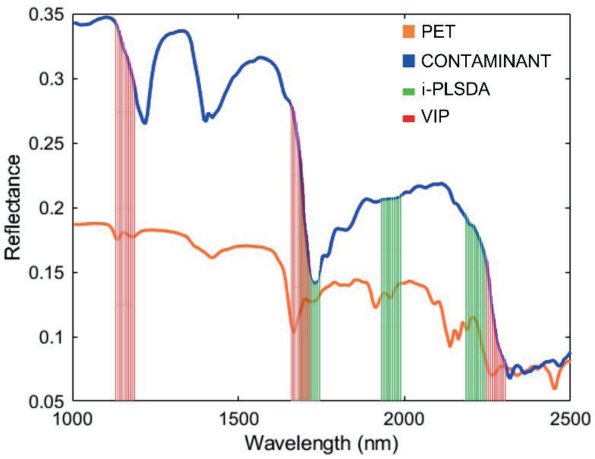

FIGURE 6: i-PLSDA prediction maps for the validation (a), test 1 (b), test 2 (c) and test 3 (d) datasets.

mostly in the validation dataset (cf. Figure 1a, 1b and 8a) and test 3 (cf. Figure 1e and 8d), except for some pixels due to border-effect (Figure 8b, 8c and 8d) and some whole samples assigned to the wrong class, marked by yellow circles in Figure 8b and 8c, i.e., test 1: 1 badly assigned sample (cf. Figure 1c and 8b) and test 2: 2 badly assigned samples (cf. Figure 1d and 8c). The classification performances, based on pixel detection, obtained by VIP (Table 2) revealed as sensitivity and specificity in calibration, cross-validation and prediction range from 0.97 to 0.99.

The classification performances, based on pixel detection, in term of efficiency in cross-validation and in prediction, were compared in Table 3. In more detail, good performances were obtained by both variable selection methods (i.e., i-PLSDA efficiency in prediction = 0.94 and VIP scores efficiency in prediction = 0.98), in fact the results were very close to the full spectrum PLS-DA model results (full spectrum PLS-DA efficiency in prediction = 1.00), considered as an ideal classification. In conclusion, the VIP scores method was the best variables reduction technique in terms of average values of sensitivity, specificity and efficiency.

4. CONCLUSIONS

In the present study, SWIR hyperspectral imaging (1000 - 2500 nm) was applied to evaluate three different PLSDA-based classification models (i.e., full spectrum PLS-DA, i-PLSDA and VIP scores) in order to develop fast and robust strategies for sensor-based sorting of PET flow stream with reference to the presence of other plastics flakes considered as contaminant. The best prediction results were provided by VIP scores method, with sensitivity, specificity and efficiency average values close to the full spectrum PLS-DA, considered as an ideal prediction model. The results demonstrated how it was possible to obtain a good identification of contaminant in a PET stream, not only considering the full investigated spectral range, but also using a reduced number of wavelength bands from 240 to 30 obtained by variable selection methods, allowing the increase of processing speed and the construction of a simpler analytical logic with reduced costs, being both necessary requirements for industrial applications.

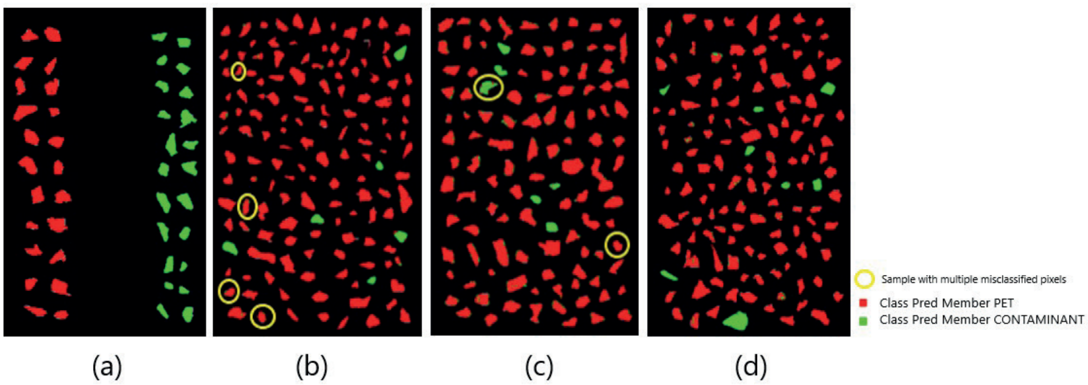

FIGURE 7: i-PLSDA prediction maps for the validation (a), test 1 (b), test 2 (c) and test 3 (d) datasets.

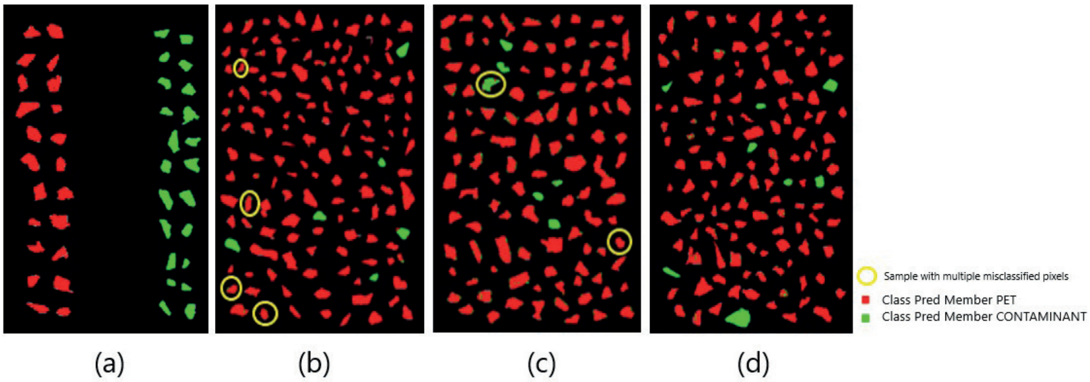

FIGURE 8: VIP prediction maps for the validation (a), test 1(b), test 2 (c) and test 3 (d) datasets.

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TABLE 2: Mean performance values of i-PLSDA and VIP classification for PET class in calibration, cross-validation and prediction phases. Used LVs: 3.

		Sensitivity	Specificity
i-PLSDA	Calibration	0.97	0.98
	Cross-validation	0.97	0.97
	Prediction	0.97	0.90
VIP	Calibration	0.99	0.98
	Cross-validation	0.99	0.98
	Prediction	0.99	0.97

TABLE 3: Classification performances in prediction and cross-validation phases in terms of mean efficiency values.

Models	Efficiency in cross-validation	Efficiency in prediction
Full spectrum PLS-DA	1.00	1.00
i-PLSDA	0.97	0.94
VIP scores	0.98	0.98

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