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A Review on Chemometric Techniques with Infrared, Raman and Laser-induced Breakdown Spectroscopy for Sorting Plastic Waste in the Recycling Industry

Authors Edward Ren Kai Neo[a,b*], Zhiquan Yeo[b], Jonathan Sze Choong Low[b], Vannessa Goodship[a], Kurt Debattista[a]

[a]*Warwick Manufacturing Group, University of Warwick, Coventry, CV4 7AL, United Kingdom*

[b]*Singapore Institute of Manufacturing Technology, 2 Fusionopolis Way, Singapore 138634, Singapore*

**Corresponding author: edward.neo@warwick.ac.uk (Email)*

Abstract

Mismanagement of plastic waste globally has resulted in a multitude of environmental issues, which could be tackled by boosting plastic recycling rates. Chemometrics has emerged as a useful tool for boosting plastic recycling rates by automating the plastic sorting and recycling process. This paper will comprehensively review the recent works applying chemometric methods to plastic waste sorting. The review begins by introducing spectroscopic methods and chemometric tools that are commonly used in the plastic chemometrics literature. The spectroscopic methods include near-infrared spectroscopy (NIR), mid-infrared spectroscopy (MIR), Raman spectroscopy and laser-induced breakdown spectroscopy (LIBS). The chemometric tools include principal component analysis (PCA), linear discriminant analysis (LDA), partial least square (PLS), k-nearest neighbors (k-NN), support vector machines (SVM), random forests (RF), artificial neural networks (ANNs), convolutional neural networks (CNNs) and K-means clustering. This review revealed four main findings. 1) The scope of plastic waste should be expanded in terms of types, contamination and degradation level to mirror the heterogeneous plastic waste received at recycling plants towards understanding potential application in the recycling industry. 2) The use of hybrid spectroscopic method could potentially overcome the limitations of each spectroscopic methods. 3) Develop an open-sourced standardized database of plastic waste spectra would help to further expand the field. 4) There is limited use of more novel machine learning tools such as deep learning for plastic sorting.

Keywords: Chemometrics; spectroscopy; hyperspectral imaging; spectral analysis; machine learning; plastic recycling

Abbreviations

ABS - Acrylonitrile Butadiene Styrene

HDPE – High Density Polyethylene

HSI - Hyperspectral Imaging

IR - Infrared

LDPE – Low Density Polyethylene

LLDPE – Linear Low Density Polyethylene

LIBS – Laser-induced Breakdown Spectroscopy

MIR – Mid Infrared

NIR – Near Infrared

PA - Polyamide

PC - Polycarbonate

PE - Polyethylene

PLA – Polylactic Acid

PMMA – Polymethyl Methacrylate

POM - Polyoxymethylene

PP – Polypropylene

PS - Polystyrene

PTFE - Polytetrafluoroethylene

PU - Polyurethane

PVC – Polyvinyl Chloride

1. Introduction

Plastic is a versatile material used for a wide range of applications, such as packaging, construction and agriculture. The demand for plastics has increased 200-fold over the past 70 years, with 381 million tons of plastics produced annually in 2015 and over 8 billion tons of total production produced to date (Geyer et al., 2017; Ritchie, 2018). Managing the resulting increase in plastics waste generated has become an increasingly critical global challenge. It has been estimated that only 9% of all the plastics ever generated have been recycled, while the vast majority were landfilled (Geyer et al., 2017).

Due to the mismanagement of plastic waste, a large amount of plastic waste has leaked into the oceans; this has been linked to a multitude of environmental issues (Jambeck et al., 2015). Plastic is known to be extremely persistent in the environment and could disrupt the marine ecosystem through pathways such as ingestion or entanglement (Ferronato and Torretta, 2019). Microplastics also accumulate in the food chain and were recently found in human placenta (Ragusa et al., 2021), which could potentially be linked to harmful effects for humans (Campanale et al., 2020). At the current rate of plastic waste management, it is projected that the weight of plastics in the ocean would exceed that of fish by 2050 (Ellen MacArthur Foundation, 2017), highlighting the severity and urgency of addressing plastic pollution, which could be addressed by boosting plastic recycling.

While plastic recycling rates have generally been trending upwards over the years, plastic recycling rates remain low globally at around 18% (OECD, 2018). The factors contributing to low recycling rate for plastics has been well studied, and includes various economic, information, technical and legislation barriers (Hopewell et al., 2009; Milios et al., 2018; Suchismita, 2017; Tesfaye and Kitaw, 2020). Proper sorting of plastic waste is one way to overcome some of the barriers. Plastic sorting has traditionally relied on a combination of manual labor and physical methods (Dodbiba and Fujita, 2004). These traditional methods utilize physical properties of plastic for sorting, such as density and electrical conductivity. More detailed reviews of these traditional methods have previously been conducted and will be out of scope of this paper (Al-Salem et al., 2009; Gundupalli et al., 2017; Malcolm Richard et al., 2011; Wang et al., 2015; Wu et al., 2013).

One drawback of physical methods is the lack of a feedback mechanism to constantly monitor the quality of plastic waste going into recycling, limiting the traceability of plastic type and quality for recycling. In recent times, solutions focusing on automated plastic sorting systems with machine learning techniques are on the rise. Some systems have approached it as an image recognition task, which is useful for identifying common plastic products like mineral water bottles (Wang et al., 2019). Other systems have explored using chemometrics, which involves the use of chemical data from spectroscopy methods (Heberger, 2008) for automatic sorting of plastic waste. Chemometrics have been widely applied towards quality control in the food (Liang et al., 2020) and pharmaceutical industries (Biancolillo and Marini, 2018), environmental modelling (Chapman et al., 2020) and forensics (Sauzier et al., 2021), but chemometric techniques have only recently gained popularity in the area of plastic waste (da Silva and Wiebeck, 2020). A broad review of various physical and chemometric-based method for municipal solid waste sorting was performed recently (Gundupalli et al., 2017). Other reviews have studied the use of chemometrics for microplastics detection with Raman (Araujo et al., 2018) and Fourier transform infrared (FTIR) spectroscopy (Veerasingam et al., 2020), but none, so far, have focused on plastic waste sorting.

This work aims to build upon the literature by comprehensively reviewing the use of chemometric method specifically for plastic waste sorting, and assessing the state of the field for application in the recycling industry. This would contribute towards helping to determine the research directions that can be undertaken to help develop a state-of-the-art plastic sorting approach which can effectively sort out recyclable polymers from post-consumer plastic waste. The surveyed literature was found using the search terms ‘plastic’, ‘recycling’ and ‘chemometrics’ or one of the spectroscopic method ‘Infrared’, ‘Raman’, ‘LIBS’ on Scopus, Web of Science and Google Scholar. Section 2 will provide a background on spectroscopic methods that can be used to obtain chemical data of plastics, and chemometric techniques to analyze the chemical data; section 3 will cover methodology applied for the literature review; sections 4 to 6 will cover specific works that have been done in this field using infrared, Raman and LIBS data respectively; section 7 will discuss and evaluate the limitations and gaps in using spectroscopic data for chemometric analysis.

2. Background

This section will first introduce some spectroscopic methods that have been widely used to obtain chemical data from plastic in a non-destructive manner. Following that, chemometric techniques that are widely used to analyze spectroscopic data of plastics will be introduced.

2.1 Spectroscopic Methods

Spectroscopy is the study of interaction between electromagnetic radiation and matter. There are broadly three types of spectroscopic methods that are commonly used for chemometrics sorting of plastic, which are infrared spectroscopy, Raman spectroscopy and laser-induced breakdown spectroscopy, which will be described in this section. A comparison of the three spectroscopic methods will be further presented in section 7.

2.1.1 Infrared Spectroscopy

Infrared (IR) spectroscopy measures the absorption or reflectance of IR radiation by chemicals or materials (Griffiths et al., 2007). The electromagnetic radiation in this region is typically associated with the rotational and vibrational frequencies of different chemical bonds within the molecules. These are termed resonant frequencies, which are absorbed by the molecules, while the other frequencies would be transmitted. Fourier transform infrared spectroscopy (FTIR) applies a mathematical technique known as Fourier transform to convert the raw time domain signals into an easily visualizable IR spectrum (Griffiths et al., 2007), which maps the IR radiation absorbed/transmitted over each frequency, thus generating a molecular fingerprint. The IR region is further divided into far-, mid- and near-IR, each containing different information. Rotational frequencies can be found within far-IR, while fundamental vibrational frequencies can be found within mid-IR and overtones of vibrational frequencies can be found within near-IR (Veerasingam et al., 2020).

2.1.2 Raman Spectroscopy

Raman spectroscopy is another technique used to study the rotational and vibrational frequencies of chemical bonds within a molecule (McCreery, 2005). When photons from a laser interact with the molecular vibrations, molecules can be excited to a higher energy level. Most of this energy will be dissipated through elastic scattering (or Rayleigh scattering), where the energy of the emitted photons is equal to the photon from the laser. Raman spectroscopy measures the wavelength of inelastically scattered photons, where the emitted photon is higher or lower in energy as compared to the photon from the laser, which can be visualized in a spectrum of intensity over wavelength. As the majority of scattered photons are elastically scattered, a light filter is often used to filter out the scattered radiation to allow better observation of the inelastically scattered photons.

2.1.3 Laser-induced Breakdown Spectroscopy

Laser-induced breakdown spectroscopy (LIBS) is an elemental analysis technique that can be used to detect the presence of all elements (Singh and Thakur, 2020). It focuses a high energy laser on a sample to vaporize and atomize a small amount of material into plasma. Characteristic radiation emitted by each of the element can be detected to confirm the presence of different elements.

2.2 Chemometric Techniques

The chemical data obtained via spectroscopic methods can be analyzed with various chemometric tools. While there are significant numbers of possible tools, this work will focus on the ones used in the chemometrics publications. A broad category of tools frequently used in chemometrics research are dimensionality reduction tools such as principal component analysis (PCA), linear discrimination analysis (LDA) and partial least squares regression (PLS). Supervised machine learning, where labelled data is used to train the model to classify the plastic from the spectrum obtained using spectroscopic methods, can also be used. These include tools like k-nearest neighbor (k-NN), support vector machines (SVM), random forest and neural networks. Limited works also explored unsupervised machine learning in the form of k-means clustering.

2.2.1 Principal Component Analysis

PCA is a dimensionality reduction tool used for multi-dimensional datasets (Jolliffe and Cadima, 2016; Smith, 2002). This statistical tool constructs new axes known as principal components that are linear combinations of the initial variables. Each principal component is constructed in a way to maximize the variance, hence capturing as much information as possible. The contribution of each principal component to the overall data variance can be visualized by the explained variance ratio. By plotting the principal components with the highest explained variance ratio, the data can easily be visualized in a two-dimensional graph. Data belonging to the same category would typically be clustered together in the plot and would be sufficiently distinct from other clusters. PCA has been used to reduce the dimension of plastic spectra before passing the principal components as input data into different classifier models (Musu et al., 2019; Yang et al., 2020; Zhu et al., 2019). A sample PCA plot for high density polyethylene (HDPE) and polyethylene terephthalate (PET) is shown in Fig 1.

Soft independent modelling by class analogy (SIMCA) is an extension of PCA used for classification. For each class, a plane or hyper-plane is constructed from the PCs, and new samples are classified based upon the distance to the plane or hyper-plane. This method is a soft classification, as it is possible for each sample to be classified into multiple classes, depending on the threshold distance for classification (Costa et al., 2017; Wienke et al., 1995).

2.2.2 Linear Discrimination Analysis

LDA is related to PCA as a dimensionality reduction tool. However, in LDA, new axes are constructed in a way that maximizes class separation (Izenman, 2008). This is done by maximizing the distance between the means of each class while minimizing the scatter of the dataset within each class. The data is then mapped to the new lower dimension axis. LDA was used to classify different plastics (Wu et al., 2020) and plastics with different types of brominated flame retardants (Stefas et al., 2019).

2.2.3 Partial Least Square

PLS regression is a statistical method that uses latent variables to study the relationship between two matrices (Haenlein and Kaplan, 2004) The latent variables are constructed in a way to find the vector in the X space that corresponds to the vector in the Y space with maximum variance. When used for classification problem, a variant called PLS discriminant analysis (PLS-DA) is used, where the Y matrix will be expressed as a dummy matrix of 1 and 0. PLS-DA is one of the more popular chemometric techniques that has been used for classifying different types of plastics (Calvini et al., 2018; Liu et al., 2019b; Pieszczek and Daszykowski, 2019; Saeki et al., 2003; Sato et al., 2002; Silva and Wiebeck, 2019).

2.2.4 k-Nearest Neighbor

k-NN is a classification algorithm which classifies based upon the identity of the k-nearest neighbors to the new observation (Altman, 1992), where k is a parameter that can be tuned. The majority class in the k nearest neighbors will determine the class of the new observation. k-NN can also be combined with PCA for datasets with large dimensions, which is illustrated in Fig 1. Costa et al., (2017) and Yang et al., (2020) used this algorithm in their works on plastic classification.

2.2.5 Support Vector Machine

SVM is a classification algorithm that constructs a decision boundary to maximize the distance between the different classes (Wang, 2005). New samples will then be classified based on the side of the decision boundary that it falls on in a non-probabilistic manner. Traditionally a binary classification algorithm, more recent advancement allows for SVM to solve multi-class classification problems (Lee et al., 2004), including for plastic classification (Musu et al., 2019; Yang et al., 2020; Yu et al., 2014; Zhu et al., 2019). SVM can also be combined with PCA for datasets with large dimensions, which is illustrated in Fig 1.

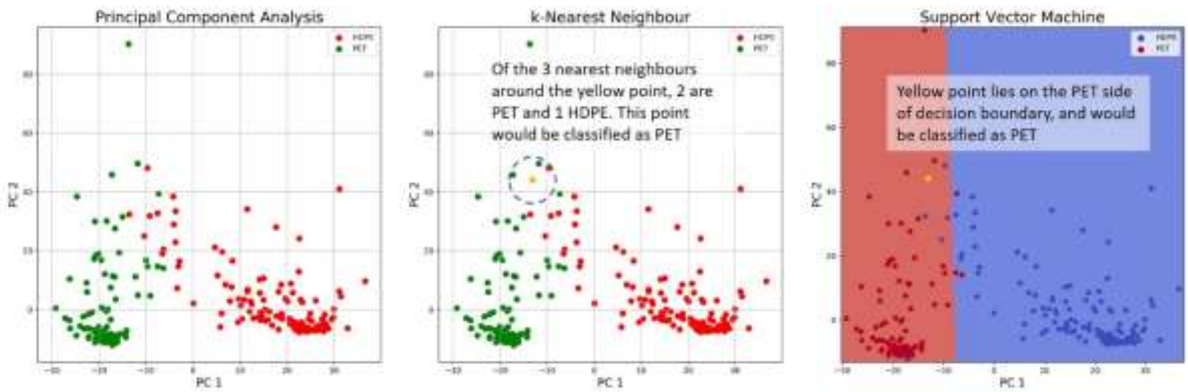

Fig 1. PCA, k-NN and SVM for chemometric analysis of plastic FTIR data. The dataset from Chabuka and Kalivas, (2020) was used to build the classifier.

2.2.6 Random Forests

Random forest is an ensemble machine learning technique built using many decision trees (Breiman, 2001). The output of the random forest algorithm is the mean of the prediction outputs of all the decision trees. Bagging is employed during the learning process, where each decision tree is built using different training data. This helps to prevent overfitting to the dataset. Random forest regression was used with LIBS to quantify the presence of toxic heavy metal elements in plastics (Liu et al., 2019c).

2.2.7 Neural Networks

Artificial neural networks (ANNs) fall under a branch of machine learning known as deep learning used for predictive modelling (Krenker et al., 2011). The inputs matrix in the input layer is mapped to the classified output using neurons in hidden layers. Each of the neurons contains a function that applies a weight to different part of the input, and together all the neurons help to learn a complex function. During the training process, the weights are usually adjusted through an iterative backpropagation process to reduce the loss via gradient descent. Variations of ANN architectures have been widely used in plastic classification (Bae et al., 2019; Boueri et al., 2011; Junjuri and Gundawar, 2020, 2019; Musu et al., 2019; Roh et al., 2017; Saeki et al., 2003; Wienke et al., 1995; Yang et al., 2020). A sample ANN architecture for classifying plastics is shown in Fig 2.

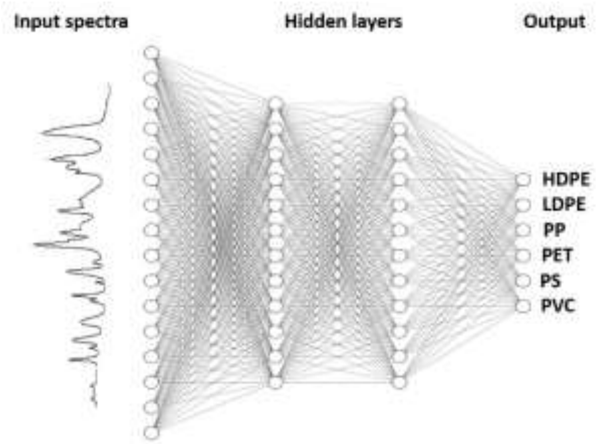

Fig 2. Sample ANN architecture for plastic classification. The image was built with NN-SVG.

Convolutional neural network (CNN) is one variant of neural network architecture developed in recent years (Lecun et al., 1998) that has been used with spectra signals (Chen & Wang, 2019; Liu et al., 2017; Ng et al., 2019, 2020; Stiebel et al., 2018; Zhang et al., 2019). CNNs consist of three types of layers – convolutional layers that extracts features from the input data, pooling layers that help to reduce the dimension, and fully connected layers that are essentially ANNs. A sample CNN architecture for classifying plastic is shown in Fig 3.

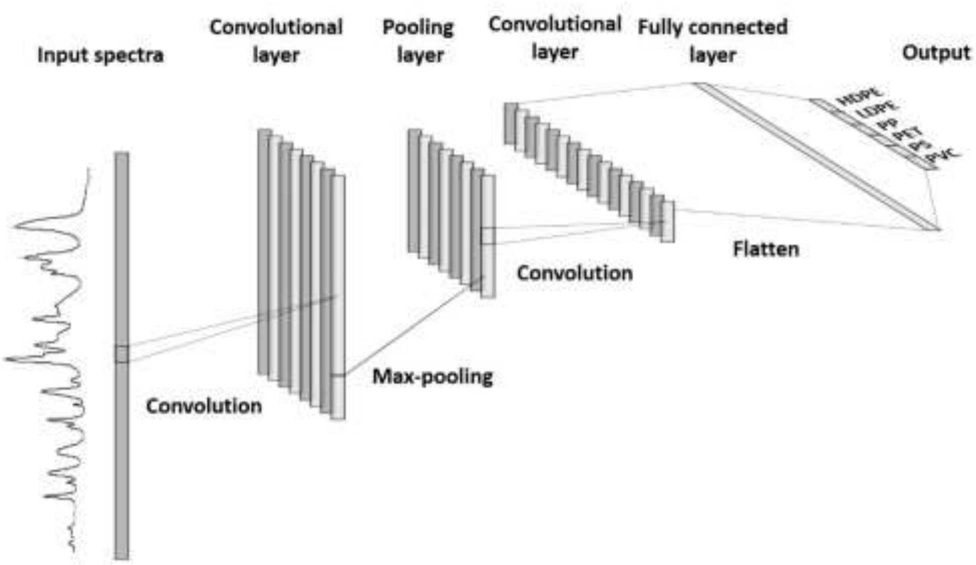

Fig 3. Sample CNN architecture for plastic classification. The image was built with NN-SVG.

2.2.8 K-means Clustering

K-means clustering is an unsupervised machine learning algorithm which groups each observation into one of k clusters based on the distance to the centroid of each cluster, where k is a hyperparameter to be tuned (Likas et al., 2003). During the learning process, the cluster centroids are first randomly defined, and each observation is assigned to the nearest centroid based on Euclidean distance. A new centroid is then defined for all the points in each cluster, and the process is repeated iteratively until convergence is reached. While K-means clustering is not meant to a classification algorithm, polymers of the same type are likely to be clustered together, which could result in good polymer sorting if each of the cluster represents a particular polymer type. K-mean clustering was used by Guo et al. (2018) to group 20 different polymer samples into each own cluster..

3. Methodology

The following sections will discuss the results and limitation from previous chemometric studies for plastic sorting. This section will outline the characteristics that will be discussed for each method.

For each study, the methodology and the dataset characteristics will be summarized. In particular, for each study we will outline:

1) Spectroscopic method – IR spectroscopy (including near IR, mid IR and hyperspectral imaging), Raman spectroscopy and LIBS.
2) Samples – the resin type of samples used in the study, which includes the six common types of high density polyethylene (HDPE), low density polyethylene (LDPE), polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS), polyvinyl chloride (PVC), and other types such as acrylonitrile butadiene styrene (ABS), polyamide (PA), polycarbonate (PC), polylactic acid (PLA), polymethyl methacrylate (PMMA), polyoxymethylene (POM), polytetrafluoroethylene (PTFE) and polyurethane (PU).
3) Hardware – the instrument and its specifications.
4) Input – the parts of the chemical data obtained from spectroscopy that is used for chemometric analysis.
5) Software – the software used to run the chemometric tool.
6) Chemometric tool – tools used to analyze spectroscopic data, including PCA, LDA, kNN, PLS, SVM, RF, neural networks.
7) Results – accuracy, precision and recall (equation 1-3) for classification, root mean square error of prediction (RMSEP) for regression.
8) Dataset characteristics – the data size, color and source of plastic samples (Table S1 to S3).

The equations used for 7 above are:

$$Accuracy = \frac{TP+TN}{TP+FP+TN+FN} \quad (1)$$

$$Precision = \frac{TP}{TP+FP} \quad (2)$$

$$Recall = \frac{TP}{TP+FN} \quad (3)$$

$$Specificity = \frac{TN}{TN+FP} \quad (4)$$

Where TP is True Positive, FP is False Positive, TN is True Negative and FN is False Negative.

Most chemometric classification studies only report the accuracy of plastic sorting, which is useful as a general indication of the model performance. However, whenever possible, this review will also derive precision and recall metrics for a more complete picture. Precision allows for understanding the level of contamination associated with the sorted polymers. However, when some studies report specificity values (equation 4) without a confusion matrix, making it impossible to derive the precision value. For those studies, the specificity metrics was reported instead. Recall allows for understanding the percentage recovery of polymers of each class. To extract the highest market value from plastic waste, precision should be prioritized to produce high quality recycled plastic, followed by recall to ensure that potentially recyclable polymers are not downcycled. However, the recall metric should also be suitably high such that a fair amount of recyclable plastic can be recovered for the process to be economically sustainable.

The summarized dataset characteristics include the plastic sample color and source. Color on plastics is due to either organic or inorganic colorants, while the source may give an indication of possible contamination and degradation. These are important factors that could affect both identification of the plastic and the recyclability of the plastic. Different colored plastic should be processed separately during the recycling process for an aesthetically pleasing recycled product (Ruj et al., 2015). Colorless or white samples have the highest market value as recycled plastics, since they can be re-dyed with any color (Gabriel and Maulana, 2018). On the other hand, black samples can only be recycled into black plastics. Contaminants found on plastics are typically volatile organic compounds (VOCs), which affects the odor quality of the recycled product (Cabanes et al., 2020; Strangl et al., 2021). Contaminants like detergent could also be linked to increased thermal degradation effects during the recycling process (Mylläri et al., 2016). Degradation of plastics are usually associated with formation of carbonyl or hydroxyl groups on the surface (Canopoli et al., 2020; Pelegrini et al., 2019) which causes changes in the resulting spectra that could result in misclassification. Degradation also results in decreased mechanical and rheological properties as compared to virgin plastic (Brouwer et al., 2020).

4. Chemometrics with IR

Among the three spectroscopic methods discussed in section 3, IR spectroscopy is the most widely applied spectroscopic method for chemometric analysis for plastic waste sorting. Broadly speaking, FTIR spectroscopy can be split into 3 ranges – near-infrared (NIR) with wavenumber from 14,000 – 4000 cm^{-1}, mid-infrared (MIR) with wavenumber from 4000 – 400 cm^{-1} and far infrared (FIR) with wavenumber from 400 – 10 cm^{-1} (Veerasingam et al., 2020). Both NIR and MIR are suitable ranges for plastic sorting.

4.1 Near Infrared

Table 1 summarizes the methodology and result of works in the literature that utilized NIR for chemometric analysis of plastic waste.

The use of NIR for chemometric analysis of plastic waste is well established, which is evident from the results (average classification accuracy ranging from 97-100% in the reported works). Early works identified ANN in combination with NIR as an effective method for sorting common post-consumer plastic waste with high accuracy (Feldhoff et al., 1997; Huth-Fehre et al., 1995; Wienke et al., 1995). Highly sensitive detectors in the NIR region by indium gallium arsenide (InGaAs) based sensors also makes it suitable for use with in-line conveyer belt systems in the recycling industry (Feldhoff et al., 1995). Much of the research direction afterwards shifted towards NIR hyperspectral imaging (HSI), which involves generating a 3D 'hypercube' with two spatial dimensions and one spectral dimension (Caporaso et al., 2018). and will be further covered below in Section 4.1.1. More recent works explored the use of portable NIR systems and found that they were as effective (Kumar et al., 2014; Rani et al., 2019; Said et al., 2020; Yang et al., 2020), which opens the door for potential deployment in decentralized sorting systems such as smart sorting bins.

NIR data has also been used to build regression models that provides good age-prediction accuracy for plastics by subjecting plastic samples to thermal-oxidative aging and extrusion cycles (Alassali et al., 2020, 2018), which could provide useful information for determining the recyclability of plastic waste.

Table 1: Summary of NIR chemometrics study for plastic waste sorting.

Reference	Samples	Hardware	Input	Software	Chemometric Tool	Accuracy	Precision	Recall	Main Misclassification
1 (Huth-Fehre et al., 1995; Wienke et al., 1995)	PE, PP, PET, PS, PVC	PolyTec X-DAP, InGaAs detector	Full NIR spectrum (825 – 1700nm)	ARTHUR MATLAB	SIMCA ANN		Median sorting purity of 98%		-
2 (Feldhoff et al., 1997)	PE, PP, PET, PS, PVC	PolyTec X-DAP, InGaAs detector	Full NIR spectrum (825 – 1700nm)	Not stated	ANN	Overall 97%			PE and PP
3 (Saeki et al., 2003)	HDPE, LDPE, LLDPE	PlaScan-SH (Opt Research Inc., Japan)	Second-derivative of full NIR spectrum (1100-2200nm)	Pirouette, NEUROSIM/L	PCR PLS ANN		RMSEP of 0.0043 RMSEP of 0.0031 RMSEP of 0.00026 PE density range 0.898-0.962 g cm^{-3}		-
4 (Zhao and Chen, 2015)	PE, PP, ABS, PMMA	Jiaoda spectrometer	First 3 Principal components for NIR spectrum (900-1700nm)	Not stated	Mahalanobis distance	88.9-100% (Overall 97%)	88.9-100% (Overall 97%)	90.9-100% (Overall 97%)	ABS predicted as PP

5 (Zhu et al., 2019)	PE, PP, PET, PS, ABS, PMMA	NIR optical fiber spectrometer with InGaAs detector	First 7 principal components for NIR spectrum (1000-1700nm)	LabVIEW	PCA-SVM	85-100% (Overall 97.5%)	87-100% (Overall 97.8%)	85 – 100% (Overall 97.5%)	PE predicted as PP
6 (Rani et al., 2019)	PE, PP, PET, PS, PVC	MicroNIR On-site	Full NIR spectrum (900-1700nm)	MicroNIR TM Pro v3.0 software	PLS-DA	Overall Accuracy: 100%			
7 (Wu et al., 2020)	PP, PS, ABS, ABS/PC blend	Ocean Optics NIR512	Part of NIR spectrum (1084-1562nm)	Python	PLS-DA PCA-LDA	99.5-100% (Overall 99.9%)	99.1-100% (Overall 99.9%)	99.5-100% (Overall 99.9%)	-
8 (Yang et al., 2020)	PE, PP, PET, PVC, PS, ABS, PC White and transparent samples	Pynect NIR-S-G1 NIR handheld spectrometer	Full NIR spectrum (900-1700nm)	Python (PyCharm)	PCA-SVM PCA-KNN PCA-ANN	100% (All) 100% (Transparent) 99.95% (White) 100% (Transparent) 99.94% (White)	100% (All) 100% (Transparent) 99.96% (White) 100% (Transparent) 99.94% (White	100% (All) 100% (Transparent) 99.95% (White) 100% (Transparent) 99.94% (White	-
9 (Said et al., 2020)	PP, PET	Miniaturized MEMS FTIR spectrometer	First 2 latent variables for NIR spectrum (1350-2500nm)	Not stated	PLS, KNN	Overall Accuracy: 100%			-

4.1.1 Near Infrared Hyperspectral Imaging

Hyperspectral imaging (HSI) in the NIR range have started to be adopted by some industries (WRAP, 2016), The imaging capabilities help to provide information about the purity of plastic samples through the spatial distribution of different spectra on the sample. Some of the NIR hyperspectral imaging technologies currently in the market includes the Specim FX17 (Specim, 2020a), KUSTAx.x MSI series (LLA Instruments, n.d.), Pika NIR series (Resonon, 2020) and INNO-SPEC RedEye (Acal Bfi, 2015a), some of which have been utilized in the reported literature (Calvini et al., 2018; Pieszczek and Daszykowski, 2019). Table 2 summarizes the methodology and result of the work in the literature that utilized HSI-NIR for chemometric analysis of plastic waste.

Initial work in this field using PCA found clear separation between two different groups of polymers such as PE and PP, PE and PET or PET and PLA (De Biasio et al., 2010; Serranti et al., 2012, 2011; Ulrici et al., 2013). More recently, 100% accuracy was obtained in classifying PE and PP using PLS-DA (Serranti et al., 2015). With an increase in scope of polymer class considered, there is a slight drop in overall results (Calvini et al., 2018; Karaca et al., 2013; Pieszczek and Daszykowski, 2019; Stiebel et al., 2018) as compared to the results shown in Table 1 with the use of NIR spectrometer, which could be partly due to poorer resolutions in HSI-NIR spectra. However, the accuracies reflect pixel accuracies, rather than sample accuracies. In most cases, the majority of pixels in the plastic sample is correctly classified, which would have likely resulted in positive identification of the sample. Furthermore, it was also found that using selected bands representing 10% of the initial input data do not compromise the overall performance (Kim and Kim, 2016). To further improve the sorting accuracy, the effect of different pre-processing methods can be explored, as it can significantly influence the final results (Galdón-Navarro et al., 2018).

More recent works have attempted to make use of the spatial information captured within HSI-NIR for more detailed chemometric analysis, such asin detecting contamination in plastics such as bromine flame retardants (Bonifazi et al., 2020; Caballero et al., 2019), identifying multi-layered polymers (Bonifazi et al., 2021; Chen et al., 2021b; Stiebel et al., 2018) and distinguishing between plastic of varying degradation levels (Chen et al., 2021a),

While the research in this field is mature, there remains some gaps in this field. Most of the work has focused on variations of the six common plastic – HDPE, LDPE, PET, PP, PS and

PVC. In reality, the mix of plastic waste would be much more heterogenous, which may affect the accuracy of chemometric models. The use of NIR in chemometric analysis of polyolefins should also be better understood, as not many works have split the class of PE in HDPE and LDPE (Calvini et al., 2018; Karaca et al., 2013; Saeki et al., 2003; Stiebel et al., 2018). Some work also reported mislabeling of some PE and PP samples (Pieszczek and Daszykowski, 2019; Zhu et al., 2019). There is also lack of an open-sourced NIR polymer database, which limits further research in this field.

Furthermore, there are glaring limitations of NIR in sorting plastic waste. One of the major drawbacks with NIR is the inability to differentiate black plastics, as electromagnetic radiation in the NIR region is strongly absorbed by black material due to its proximity to the visible light range (Becker et al., 2017). Furthermore, NIR is composed of overtones and combination bands of different functional groups such as C-H, N-H and O-H, resulting in weaker spectral features that may present difficulties towards unique identification (Vázquez-Guardado et al., 2015). In order to manage the heterogenous plastic waste at recycling facilities, NIR data can be supplemented with data from other non-destructive spectroscopic sets like MIR, Raman and LIBS, which will be covered in later sections.

Table 2: Summary of HSI-NIR chemometrics study for plastic waste sorting.

Reference	Samples	Hardware	Input	Software	Chemometric Tool	Accuracy	Precision	Recall	Main misclassification
1 (De Biasio et al., 2010)	PE, PP	NIR scanner from Titech	HSI-NIR (1000-2500nm)	Not stated	PCA	PE and PP can be distinguished, different PP products can be distinguished as well.			-
2 (Serranti et al., 2011)	PE, PET	Specim NIR Spectral Camera with InGaAs detector	HSI-NIR (1000-1700nm)	Spectral Scanner v2.3	PCA	PE and PP can be distinguished from other contaminants (aluminum, wood, foam)			-
3 (Serranti et al., 2012)	PE, PP	Specim NIR Spectral Camera with InGaAs detector	HSI-NIR (1000-1700nm)	Spectral Scanner v2.3	PLS-DA	Not stated	94-95%	94-95%	
4 (Karaca et al., 2013)	HDPE, LDPE, PP, PET, PVC, PS	SWIR Camera	HSI-NIR (1000-2500nm)	Self-developed	SVM	93.5-96.9%	Not stated	Not stated	LDPE predicted as HDPE
5 (Ulrici et al., 2013)	PET, PLA	Specim ImSpector N17	HSI-NIR (1000-1700nm)	Not stated	PLS-DA	98.7-100%	Not stated	Not stated	-

6 (Bonifazi et al., 2014)	PE, PP, PET, PS, PVC	Specim ImSpector N17	HSI-NIR (1000-1700nm)	MATLAB	PLS-DA	Not stated	99.8-100%	100%	-
7 (Serranti et al., 2015)	PE, PP	Specim ImSpector N17	HSI-NIR (1000-1700nm)	MATLAB	PLS-DA	100%	100%	100%	-
8 (Calvini et al., 2018)	HDPE, LDPE, PP, PET, PVC, PS, ABS, PLA	HSI Camera KUSTA1.9M SI, LLA Instruments with InGaAs detector Zeiss f/2.4, 10 mm optical lens	HSI-NIR (1330 – 1900nm)	MATLAB	Soft PLS-DA	98.4%	Specificity: 97.4-100%	92.1-100%	
9 (Stiebel et al., 2018)	HDPE, LDPE, PP, PET, PVC, PS, ABS and mixed samples	hyperspectral NIR-camera	HSI-NIR (900-1700nm)	Python (Tensorflo w/Keras)	CNN (Hypnet)	92%	Not stated	Not stated	
10 (Pieszczek and Daszykowski, 2019)	HDPE, PP, PS, PET, ABS	Specim FX17e camera with InGaAs detector	HSI-NIR (1000-1700nm)	MATLAB	OC-PLS	Not stated	Specificity: 99.5-99.9%	93.4-98.6%	PP predicted as HDPE

11 (Serranti et al., 2020)	HDPE, PP White, red, orange, yellow green, blue samples	Specim ImSpector N25E imaging spectrograph, ImSpector V10E VIS0BUR canera	HSI-NIR (1000-2500nm)	MATLAB	PLS-DA	Not stated	<u>Type class</u> 100% <u>Color class</u> 92.6-100%	<u>Type class</u> 100% <u>Color class</u> 98.9-100%	-
12 (Chen et al., 2021a)	PE, PP, PET, PS Varying degradation	Helios-G2-320 NIR sensor from EVK DI Kerschhaggl GmbH	HSI-NIR (930-1700nm)	Python Scikit-learn	PLS-DA	Postconsumer plastic: 99 – 99.8% Landfill plastic: 89.8 – 99.5% Marine plastic: 75.5 – 90.1%	<u>Not stated</u>	<u>Not stated</u>	-

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4.2 Mid Infrared

The MIR region contains fundamental vibrational bands with distinct spectral features and is also known as the molecular fingerprint region. Since the MIR region is sufficiently distinct from the visible light region, it is not affected by black plastics (Becker et al., 2017; Signoret et al., 2020). In addition, MIR spectroscopy is less affected by surface morphology and color of the plastic sample as compared to NIR spectroscopy (Vázquez-Guardado et al., 2015). Despite the above listed advantages that MIR has over NIR, the use of this technology in plastic recycling applications has been limited by the spectral acquisition speed with less sensitive photodetectors, two of the most commonly used being deuterated triglycine sulfate and mercury cadmium telluride (Kempfert et al., 2001). Becker et al., (2017) managed to employ a photon-up conversion technique to convert MIR photons to higher energy NIR signal, which can be picked up using a more sensitive InGaAs sensor, which improves the economic viability.

Table 3 summarizes the methodology and result of two works in the literature that utilized MIR for chemometric analysis of plastic waste. Both studies suggest 100% sorting accuracy, but a further study in this field would be needed to better understand the effectiveness of MIR technology for plastic sorting. This includes broadening the types of chemometric tools and plastic samples used. Some works have begun building up MIR spectral characteristics for polyolefins and styrenic polymers towards potential industrial application (Signoret et al., 2019a, 2019b). There are also open-sourced MIR polymer data that can be used for further chemometric studies (Baskaran and Sathiavelu, 2020; Chabuka and Kalivas, 2020; Cowger et al., 2021).

4.2.1 Mid Infrared Hyperspectral Imaging

Recent technological improvements have led to MIR-HSI being introduced into the market, allowing for potential application in an industrial setting (Signoret et al., 2019a, 2019b). Some of the products on the market now includes the Specim FX50 (Specim, 2020b) and INNO-SPEC BlackEye (Acal Bfi, 2015b). MIR-HSI was recently explored via cautious machine learning method, such that samples with high uncertainty in the prediction are rejected. The study method was employed to sort styrenic polymers and polyolefins with higher purity (Jacquin et al., 2021).

Table 3: Summary of MIR chemometrics study for plastic waste sorting.

Reference	Samples	Hardware	Input	Software	Chemometric Tool	Accuracy	Precision	Recall
1 (Kassouf et al., 2014)	HDPE, LDPE, PP, PET, PS, PLA	Bruker ATR FTIR	Full MIR spectrum (600-4000cm^{-1})	MATLAB	ICA	Plastics can be discriminated from each other		
2 (Bae et al., 2019)	PP, PET, PS	Bruker ATR FTIR	Extracted peaks from MIR spectrum (600-2000cm^{-1})	WEKA 3.8	RBFNN	99-100%	Not stated	Not stated

5. Chemometrics with Raman

Raman spectroscopy is the second of the three spectroscopic methods introduced in section 3 that could potentially be combined with chemometric tool towards plastic waste sorting. Raman has emerged as a technique that overcomes the shortcomings of NIR, such as inability to handle black plastics and poor spectral resolution in a rapid fashion (Chen et al., 2017; Musu et al., 2019; Tsuchida et al., 2009). Furthermore, Raman spectroscopy is often described as a complementary technique to MIR, as vibrational modes that are IR active are often not Raman active, and vice versa. However, the use of Raman spectroscopy for plastic identification is much less widespread when compared to IR, due to background fluorescence which can often overshadow certain peaks of interest (Araujo et al., 2018; Dong et al., 2020). Table 4 summarizes the methodology and result of works in the literature that utilized Raman spectroscopy for chemometric analysis of plastic waste.

Florestan et al. (1994) first identified the potential for Raman spectroscopy to be used to distinctly identify HDPE, PP, PET and PVC through comparison to a reference library. The authors also found that PP with fillers could be differentiated from pure PP from the Raman spectra. Allen et al. (1999) later expanded the polymer scope to include LDPE and PS, but could not achieve good overall accuracy. With the rise in popularity of machine learning models, more recent works have demonstrated the potential of chemometric tools like neural networks and SVM for analysis of Raman data for qualitative analysis of plastic, achieving between 94-100% accuracy in classifying plastic (Chen et al., 2017; Musu et al., 2019; Roh et al., 2017; Tsuchida et al., 2009).

Chemometric analysis of Raman spectra has also been shown to be effective in sorting polyethylene of different densities, as the intensities of CH_2 scissoring and wagging mode in the Raman spectrum are sensitive to the crystallinity of PE (Sato et al., 2002). Allen et al. (1999) first demonstrated this with 100% correct identification of both HDPE and LDPE in his work, even while the identification of other polymers was not as effective. Other work demonstrates the quantitative analysis of the density through PLS regression models, allowing for finer distinction between HDPE, LDPE and LLDPE (Sato et al., 2002) or different HDPE/LDPE blends (Silva and Wiebeck, 2019).

Despite the demonstrated potential, there are still some gaps that need to be addressed. Most of these works extracted specific peaks for the polymer of interest to build the classification model (Musu et al., 2019; Roh et al., 2017; Tsuchida et al., 2009). This may limit the potential use in a potential industry application, where a much wider variety of polymer is encountered. Polymers that were not included in the training data could be falsely labeled, resulting in contamination that affects the quality of recycled plastic. Furthermore, no further information about the quality of the plastics, such as presence of additives and contamination, can be obtained from only studying the extracted peaks. While colored pigments can result in additional peaks or broad fluorescence bands (Florestan et al., 1994; Marica et al., 2019), the effect of different colored plastics on the performance of chemometric analysis with Raman spectra is also not well studied, as most of the works focused on plastic samples with largely homogenous colors (Chen et al., 2017; Roh et al., 2017) (Table S2).

Further work in this field should focus on broadening the scope of Raman spectra to a wider variety of polymer samples and colors that more closely resembles heterogenous plastic waste received at a recycling plant. There are currently several open-sourced Raman databases that could be utilized for such chemometric studies (Cowger et al., 2021; Dong et al., 2020; Munno et al., 2020). The potential for Raman spectra to be used with chemometric analysis for determining contamination and degradation levels within polymer samples have also not been studied.

Table 4: Summary of Raman chemometrics study for plastic waste sorting.

Reference	Samples	Hardware	Input	Software	Chemometric Tool	Accuracy	Precision	Recall
1 (Florestan et al., 1994)	HDPE, PP, PET, PVC	Bruker IFS 66 spectrometer with 300 mW YAG laser	Full Spectrum (400-4000cm^{-1})	In-built sofrware	Library searching	Not stated		
2 (Allen et al., 1999)	HDPE, LDPE, PP, PET, PVC, PS	Spex Raman Spectrometer with CCD, 514.5 nm laser	Full Spectrum (850-1800cm^{-1})	SpectraMax (in-built software)	PCA-kNN Library searching	100% 38 – 100% (Overall 87%)	Not stated	Not stated
3 (Sato et al., 2002)	HDPE, LDPE, LLDPE	JASCO NRS 2001 Raman spectrometer with CCD, 514.5 nm laser	Full Spectrum (600-1800cm^{-1})	Unscrambler	PLS	RMSEP of 0.0015 for PE density (range from 0.918 to 0.964 g/cm^3)		
4 (Tsuchida et al., 2009)	PP, PS, ABS	Homemade Raman Apparatus with CCD, 785 nm laser diode	Extracted peaks from full spectrum (300-3500cm^{-1})	R	Multivariate Analysis	Overall Accuracy: 94%	Not stated	Not stated
5 (Chen et al., 2017)	PE, PP, PET, PVC, PS PMMA, POM	LabRAM HR Evolution microscopic confocal	First seven principal components of full spectrum	Not stated	SVM	Overall Accuracy: 100%		

		Raman spectrometer, 532 nm laser	(200-2400cm^{-1})					
6 (Roh et al., 2017)	PP, PET, PS	Not stated	Extracted peaks from full spectrum (200-3000cm^{-1})	Not stated	FRBFNN	Overall Accuracy: 95%	Not stated	Not stated
7 (Musu et al., 2019)	PP, PS, ABS	Homemade Raman Apparatus with CCD, 785 nm laser diode	Extracted peaks from full spectrum (100-3300cm^{-1})	R (e1071 library) Python (TensorFlow /Keras)	PCA-SVM ANN	Overall Accuracy: 100%		
8 (Silva and Wiebeck, 2019)	HDPE/LDPE blends	Confocal Raman Microscope Alpha300 R, 532nm laser	Extracted peaks from full spectrum (210-3875cm^{-1})	MATLAB	PLS	RMSEP of 4.062 wt% of LDPE (range from 0 to 100%)		

6. Chemometrics with LIBS

LIBS is the last of the three spectroscopic methods introduced in section 3. When compared to IR and Raman, LIBS is a relatively newer spectroscopic method that has, in recent years, been used for chemometric analysis of plastic waste in a laboratory setting. (Liu et al., 2019a; Zeng et al., 2021). Since LIBS reflects the elemental composition of the sample, it does not suffer from the drawback of insensitivity to specific chemical bonds with IR or Raman spectroscopy. The relative intensity of spectral lines within a LIBS spectrum is indicative of the elemental ratio within the polymers, which can be used to differentiate different types of polymers. The ability to detect different types of elements broaden the range of plastics that can be identified, including polymers such as PTFE, PU, PA, PMMA and POM that were hardly or never explored using Raman and IR spectroscopy. Table 5 summarizes the methodology and result of works in the literature that utilized LIBS for chemometric analysis of plastic waste.

LIBS was realized as a technique that could be used for sorting common post-consumer plastics (HDPE, LDPE, PET, PP, PS, PVC) as each polymer had different C/H ratios using a simple calibration curve (Anzano et al., 2008; Gondal and Siddiqui, 2007). Some mixed success was subsequently achieved in sorting via LIBS data through Euclidean distance comparison to a reference library (Anzano et al., 2010). Banaee and Tavassoli (2012) later achieved good overall accuracy of 99% with the common post-consumer plastics using statistical methods. Since then, more recent works focused on expanding the scope of plastic for classification with various more modern machine learning methods. LIBS has also been shown to distinguish between different polymer samples of the same resin type (Guo et al., 2018; Tang et al., 2018; Yan et al., 2021), highlighting the potential in precise sorting of plastic waste. Most recently, a CNN-based approach was explored, which was found to out perform other machine learning models like ANN, SVM and kNN (Peng et al., 2021).

The studies using chemometrics with LIBS show good overall results, with 13 out of the 16 reviewed works reporting average accuracies of over 95%. The works that report high accuracies across a wide range of polymer types employed either features selection methods such as variable importance (Liu et al., 2019b) or adjusted spectral weightings (Tang et al., 2018; Yu et al., 2014). These techniques allow for accentuating weak spectral features such as CN, C_2 and O emission lines (Yu et al., 2014).

As an elemental analysis technique, chemometric analysis of LIBS can also be used to detect the presence of inorganic elements that could be linked to different additives, such as metallic-based colorants or fillers (Ángel Aguirre et al., 2013; Boueri et al., 2011; Godoi et al., 2011; Liu et al., 2019c). The chemometric methods used include ANN, RF, PLS-DA, SIMCA and KNN. Atomic spectral emissions associated with halogens like Cl and Br lie in a less sensitive region in the spectra, making it hard to observe without more specialized equipment. However, LIBS has also been used to some success to detect presence of brominated flame retardants (Stefas et al., 2019) and chlorine containing polymers (Huber et al., 2014; Vahid Dastjerdi et al., 2018).

Despite the promising results, there are some limitations associated with LIBS as an elemental analysis technique since information on the molecular structure is lost. Hence, polymers with similar elemental compositions could be hard to distinguish, such as PS and ABS (Costa et al., 2017) or PS and PC (Tang et al., 2018). While is an absence of oxygen in PS, the difficulty in sorting PC and PS was attributed to the similar relative content of C and H in both polymers, and the fact that experiments were ran in open air, where oxygen from the atmosphere interfered with the spectrum results. Supplementing LIBS data with IR or Raman spectroscopic data could potentially help to address this limitation. There could be some challenge with distinguishing between HDPE and LDPE as well, since both polymers are chemically similar. Some works have found success in this area (Costa and Pereira, 2020; Junjuri and Gundawar, 2020, 2019; Liu et al., 2019b) which could be due to the difference in additives used in both types of polymers (Arias et al., 2009).

In addition, some gaps remain in the literature. Firstly, a large majority of the works used very limited samples, often just using spectra taken from different locations of the same sample for each polymer type (Table S3). However, polymers of the same resin type from different suppliers can differ in the LIBS spectra due to presence of different additives (Peng et al., 2021). A more comprehensive study with a larger sample size would provide higher confidence that the results would generalize well to an industry. Secondly, despite the shift towards the use of LIBS for analyzing post-consumer plastics in recent years, there is a lack of open-sourced LIBS database, which limits further work in the field. Lastly, while LIBS has been used to detect the presence of metal contaminants, the potential application in predicting degradation levels are not well understood. Since degradation is associated with formation of oxygenated groups like carbonyl and hydroxyl, the O/C emission lines ratio could be a good indication of degradation levels.

Table 5. Summary of LIBS chemometrics study for plastic waste sorting.

Reference	Samples	Hardware	Spectral Lines	Software	Chemometric Tool	Accuracy	Precision	Recall	Main misclassification
1 (Anzano et al., 2010)	PE, PP, PET, PS	Q switched Nd: YAG Laser at 532nm, ICCD	Full Spectrum	Microsoft Excel	Euclidean Distance	67 – 100% (Overall 87%)	Not stated	Not stated	
2 (Grégoire et al., 2011)	PE, PP, PS, PA, PC	fourth-harmonic Nd:YAG laser, ICCD	C, H, N, O, CN, C_2	AnaLIBS (IVEA)	PCA	Polymers can be distinguished from each other			
3 (Boueri et al., 2011)	PE, PP, PVC, PTFE, POM, PA, PC, PMMA	Quadrupled Nd :YAG pulsed laser 266nm, ICCD	C, H, N, O, F, Cl Na, Mg, K, Ca, Ti CN	In-built software	ANN	81 – 100% (Overall 96%)	Not stated	Not stated	
4 (Banaee and Tavassoli, 2012)	HDPE, LPDE, PP, PET, PS, PVC	Q switched Nd: YAG Laser at 1064nm, ICCD	C, H, N, O, Cl, CN, C_2	SPSS 17.0	Discriminant Function Analysis	94 – 100% (Average 99%)	96.2-100% (Average 98.8%)	94 – 100% (Average 99%)	PP predicted as HDPE and LDPE
5 (Yu et al., 2014)	PE, PP, PVC, PS, ABS, PTFE, PA, PC, PMMA, PU, POM	Q switched Nd: YAG Laser at 532nm, ICCD	C, H, N, O, F, Cl Na, Mg, K, Ca, Ti CN, C_2	MATLAB	SVM, with adjusted spectral weightings	Overall accuracy: 100%			

6 (Costa et al., 2017)	PE, PP, ABS/PS, PA, PC	Nd:YAG Laser, CCD spectrometer	C, H, N, O	Aurora Software	KNN	91 – 100% (Overall 98%)	97 – 98% (Overall 97%)	91 – 100% (Overall 98%)
					SIMCA	89 – 96% (Overall 92%)	91 – 93% (Overall 92%)	89 – 96% (Overall 92%)
7 (Roh et al., 2018)	PP, PS, ABS	Not stated	Extracted features using PCA and ICA	Not stated	RBFNN	95.83%	Not stated	Not stated
8 (Vahid Dastjerdi et al., 2018)	PVC and others (PE, PP, PS, PMMA)	Q switched Nd: YAG Laser at 1064nm	C, N, C_2	MATLAB	SVM	90.5% (Separating PVC from other polymers)	Not stated	Not stated
9 (Guo et al., 2018)	PE, PP, PVC, PS, ABS, PTFE, PA, PC, PMMA, PU, POM	Q switched Nd: YAG Laser at 532nm, ICCD	C, H, O CN	MATLAB	K-means clustering	99.6%	Not stated	Not stated
10 (Tang et al., 2018)	PE, PP, PVC, PS, ABS, PTFE, PA, PC, PMMA, PU, POM	Q switched Nd: YAG Laser at 532nm, ICCD	C, H, N, O CN, C_2	MATLAB	SOM with adjusted spectral weightings, K-means	96 – 100% (Overall: 99%)	Not stated	Not stated

11 (Stefas et al., 2019)	ABS (with different additives)	Q switched Nd: YAG Laser	Full spectrum	Python Scikit-learn	LDA		Overall accuracy: 100%		
12 (Junjuri et al., 2019)	HDPE, LDPE, PP, PET, PS ABS, PC, HIPS,	Q switched Nd: YAG Laser at 532nm, ICCD	Full spectrum	In-built Labview programme	PLS-DA	87.2 – 97.2% (Overall 93.3%)	Not stated	Not stated	
13 (Liu et al., 2019b)	HDPE, LDPE, PP, PVC, PS ABS, PTFE, PC, PMMA, PU, POM	Solid-state Q-switched laser at 1064 nm	18 latent variables selected from full spectrum	MATLAB	PLS-DA	99.55%	Not stated	Not stated	
14 (Junjuri and Gundawar, 2019)	HDPE, LDPE, PP, PET, PS	Ti:Sapphire laser System at 800nm, ICCD	C, H, N, O Na, Mg, K, Ti CN, C_2, NH	MATLAB	ANN	97.8-100% (Overall 99.3%)	97.8-100% (Overall 99.3%)	97.8-100% (Overall 99.3%)	LDPE predicted as HDPE
15 (Junjuri and Gundawar, 2020)	HDPE, LDPE, PP, PET, PS ABS, PC, HIPS,	Q switched Nd: YAG Laser at 532nm, ICCD	C, H, N, O Na, Mg, K, Ca, Ti CN, C_2, NH	In-built Labview programme	ANN with feature selection	95.1 – 99% (Overall 97%)	Not stated	Not stated	
16 (Yan et al., 2021)	PE, PP, PVC, PS, ABS, PTFE,	Q switched Nd: YAG Laser at	20 PCs from full spectrum	MATLAB	PCA-kNN	92.1 – 100% (Overall 99.6%)	Not stated	Not stated	

	PA, PC, PMMA, PU, POM	532nm, ICCD						
17 (Peng et al., 2021)	PVC, ABS, PA, PMMA	Q switched Nd: YAG Laser at 532nm, ICCD	Full spectrum	Not stated	CNN (ResNet)	100%	100%	100%

7. Discussion

The review of relevant literature revealed the following gaps in the field: 1) scope of plastic covered, 2) hybrid spectroscopic methods, 3) open-sourced database and 4) deep learning methods. They will be covered in detail from sections 7.1-7.4.

7.1 Scope of Plastic Covered

The type of plastic considered in current studies is limited largely to some of the most common materials found in post-consumer plastic waste. Furthermore, some studies typically focus on just separating between very specific choices of plastic type. In reality, post-consumer plastic waste can also contain some less common polymers such as natural polymers and specialized engineering polymers. Exclusion of these polymers from the dataset may result in misclassification into potentially recyclable polymer classes, lowering the quality of the recycled plastic. Hence, less common plastics should also be included in future studies in order to build a more robust chemometric model for dealing with heterogenous polymer mix.

Further sorting of plastic types based on quality characteristics like contamination and degradation level, which are important considerations for the recyclability, have also not been well studied. These characteristics would be especially important for widely recyclable plastics like HDPE, PET and PP. Some preliminary works suggests that LIBS could be used to detect the presence of additives and contaminants in plastic waste, such as chlorine containing polymers (Huber et al., 2014), heavy metals (Costa and Pereira, 2020; Godoi et al., 2011) and brominated flame retardants (Stefas et al., 2019). Degradation typically results in formation of carbonyl or hydroxyl groups, leading to increased O/C ratio which can be picked up using NIR or MIR (Alassali et al., 2020, 2018; Dong et al., 2020).

7.2 Hybrid Spectroscopic Methods

While NIR is the predominant spectroscopic method used in the recycling industry today, other discussed spectroscopic methods (MIR, Raman, LIBS) have shown good potential as well, with most of the reviewed works reporting similar accuracies (well above 95%) to NIR studies. Since most of the spectra for different plastics are distinctly different, these results come as no surprise. Each of the methods have their own benefits and drawbacks, which are summarised in Table 6. NIR is the cheapest spectroscopic method of the four, but suffers in spectra resolution and dealing with black plastics (Beigbeder et al., 2013). On the flipside, MIR is not limited by black plastics, but has a much slower speed of spectrum acquisition (Kassouf et al., 2014). For IR spectroscopy, the presence of water can affect the IR spectrum due to strong absorption of IR radiation by O-H bonds. This effect is more pronounced in MIR than in NIR (Pasquini, 2018), where the O-H peaks could completely overlay other peaks of interest, most notably characteristic C-H peaks for different polymers (Primpke et al., 2020). In addition, sorting between HDPE and LDPE using IR spectroscopy could be potentially problematic, requiring some pre-processing such as using second derivatives before the difference in spectral features can be discerned (Saeki et al., 2003). The difference in spectral features between HDPE and LDPE are more distinct with Raman spectroscopy (Allen et al., 1999), but the method suffers from low sensitivity and interference of fluorescence (Dong et al., 2020). LIBS has the potential to identify the largest scope of plastics as compared to other three method, while also providing information on metallic contaminants (Ángel Aguirre et al., 2013; Liu et al., 2019c). However, LIBS spectra do not contain information about the molecular structure, and may struggle in distinguishing polymers with similar chemical formula (Costa et al., 2017).

Table 6. Comparison of different spectroscopic methods

Spectroscopic Method	**Advantages**	**Disadvantages**	**Cost (Portable options)**
NIR	• Rapid and cost-effective • Well-researched	• Weak spectral features • Unable to identify black plastics • Spectra affected by presence of water	NIR Spectrometer Ocean Insight NIRQuest - $17,000 (Ocean Insights, n.d.) StellarCASE-NIR - $20,000 (StellarNet Inc, n.d.) NIR HSI Specim FX17 – $42,500 (Stuart et al., 2020)
MIR	• Intense spectral features • Not limited by black plastics	• Slow spectral acquisition • Spectra strongly affected by presence of water	MIR HSI Specim FX50 - $200,000 (Stuart et al., 2020)
Raman	• Able to distinguish PE of different densities	• Strongly affected by fluorescence • Low sensitivity	Ocean Insight QE Pro Raman Series Spectrometers - $15,000 (Ocean Insights, n.d.) StellarCASE-Raman - $20,000 (StellarNet Inc, n.d.)

LIBS	• Applicable to large polymer scope • Able to identify metallic contaminants	• Struggle in distinguishing polymers with similar chemical formula	StellarCASE-LIBS - $30,000 (StellarNet Inc, n.d.)

Table 6 shows that there is not an ideal single method for all possible plastic waste fractions with each having pros and cons. This points to potential benefits of combining different methods for plastic waste sorting, but this has not been well studied in the literature. In particular, LIBS and Raman spectroscopy share some synergies in terms of instrumentation, since both methods involve focusing a laser beam onto the sample, but at different energy requirements. (Jolivet et al., 2019). Shameem et al., (2017) studied a hybrid LIBS-Raman system with PE, PP, PET and PS, and found that both methods offer complementary information. Raman spectroscopy resulted in a clearer separation of different transparent polymer types but the colored plastic did not form any clear cluster. On the other hand, LIBS data formed distinct clusters for each of the different plastic types regardless of the color, but the data were a lot less distinct on a PCA plot. In a related area, Ng et al., (2019) studied the use of NIR and MIR both separately and in combination for predicting soil properties. The model built using NIR data was found to perform the worst, while the model built using MIR data perform at a similar level to the model built using combined NIR and MIR data, which might point to the redundancy of NIR data in a hybrid system. Therefore, future studies can focus on performance of hybrid spectroscopic chemometric systems. Developing a unified, open-sourced database that contains different spectra of the same sample would greatly benefit exploration of this research direction.

7.3 Development of Open-Sourced Database

There are currently existing attempts at building an open-sourced database (Cowger et al., 2021; Munno et al., 2020). However, in order to realize further developments in the field, namely expanding of plastic scope and hybrid spectroscopic methods as discussed above, the information captured in current databases should be expanded, or another standardized open-sourced database should be developed. This database could contain spectra data of polymers as a pristine stage, and after simulated aging and contamination (Chabuka and Kalivas, 2020; Jung et al., 2018; Munno et al., 2020). The difference in spectral features can then be better captured towards building a chemometric tool for plastic waste sorting that can provide information on both plastic type and quality. Furthermore, spectra from each of the discussed spectroscopic methods (NIR, MIR, Raman and LIBS) should be captured for each polymer sample. This allows for potential study to understand whether the use of hybrid spectroscopic methods can offer better performance in the chemometric analysis. The development of an open-sourced database can also help to facilitate further exploration of deep learning as chemometric tool (further discussed in the section below), which relies on large amount of data for training.

7.4 Deep Learning as Chemometric Tool

Deep learning techniques are considered state-of-the art in many tasks as it allows for learning of more intricate features as compared to traditional machine learning models (LeCun et al., 2015). Several works in the literature have explored the use of neural networks as chemometric tool, but most of the network architectures explored have been basic three-layered ANNs. In recent years, the field of deep learning has expanded rapidly with different variants of other neural network architectures like convolutional neural networks (CNN) and recurrent neural networks (RNN) and generative adversarial networks (GAN) (Wang et al., 2020). In particular, several works have explored the combination of more novel neural network architectures with different spectral data in other areas (Chen & Wang, 2019; Liu et al., 2017; Ng et al., 2019, 2020; Peng et al., 2021; Stiebel et al., 2018; Zhang et al., 2019), but only Stiebel et al. (2018) and Peng et al. (2021) have applied it to plastic sorting. Due to the feature extraction nature of the network architecture, CNN has been shown to perform well even without any pre-processing of spectral data (Liu et al., 2017), which reduces the model computation time needed as compared to other chemometric tools. When applied to classification of the same polymer LIBS dataset, deep learning models were found to outperform machine learning models like ANN, SVM and kNN (Peng et al., 2021), which supports the case that the use of deep learning as chemometric techniques for plastic sorting that should be further explored.

8. Conclusion

Tackling plastic pollution remains one of the key challenges of the 21st century. A lot of research has been done to help transition the plastic economy to a circular economy, but many barriers still remain today. In the increasingly digital and fast-moving world, an automated system built upon chemometrics has shown great potential in helping to boost recycling rates by improving the sorting process. This review presented a comprehensive overview of the recent works combining the following non-destructive spectroscopic methods - Infrared spectroscopy, Raman spectroscopy and Laser-induced breakdown spectroscopy with chemometric tools like principal component analysis, partial least square regression, k-nearest neighbors, support vector machine and neural networks. Through this review, it can be concluded that chemometrics combined with non-destructive spectroscopic methods show good potential in sorting plastics. In an industrial setting, the implementation of chemometrics have started with near infrared, but the suitability of other spectroscopic methods can be further tested. The review also reveals that there is potential for further work in this field to derive further insights from chemical data. Broadly speaking, these include 1) the need to incorporate other less common polymers or polymers of varying contamination and degradation levels in training chemometric models, 2) the use of hybrid spectroscopic methods as input data to overcome the limitations of each of the spectroscopic method, 3) building a standardised dataset for plastic waste and 4) exploring deep neural networks. By expanding the literature in these directions, the authors hope that industries will be able to optimize the recycling process by capturing the maximum value out of plastic waste and transition into a circular economy.

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