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Vis/NIR and FTIR spectroscopy supported by machine learning techniques to distinguish pure from impure Iranian rice varieties

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A b s t r a c t. Rice is an annual plant from the family of Oryzeae, provides the main food for about 2.5 billion people. The quality of this product is under the influence of various factors. Quality control and adulteration detection are among the main issues in the rice industry for which, various methods have been developed. Some of these methods are costly or with low accuracy. Therefore, this study aimed to investigate and detect adulteration with spectroscopic devices and chemometric methods as well as neural network approach. The results of this study indicated the highest accuracy (100%) in the detection of authentic rice for Fourier-transform infrared combined with C-support vector machine (linear and polynomial functions) and visible–near–infrared device with quadratic discriminant analysis, multivariate discriminant analysis, Bayesian, and Decision Tree. The lowest accuracy was also related to support vector machine method with Sigmoid function for both devices. Principal component analysis method also provided very high accuracy for both devices (accuracy of 100% for visible–near–infrared and 99% for Fourier-transform infrared).

K e y w o r d s: spectroscopy, authenticity verification, rice quality control, machine learning algorithms

1. INTRODUCTION

Rice is an annual weed plant with distributed roots, robust, and white-colored from the family of Oryza, belonging to the Oryzeae tribe (Van Nguyen and Ferrero, 2006). This product is the main food of about 2.5 billion people supplying 25% of their required energy (Qamar *et al.*, 2013). Thailand, Myanmar, Vietnam, Laos, Philippines, Indonesia, Pakistan, The US, India, Japan, Italy, China, Egypt, Brazil, Mexico, Cuba, and Australia are among the main producer of rice. Hashemi, Tarom, and Sadri are among the best and highest-quality cultivars of Iranian-native rice (Rasooli Sharabiani and Khorramifar, 2022; Yinian *et al.,* 2022; Hu *et al.,* 2023).

Thanks to its high quality (delicacy, color, shape, taste, and odor), fragrant rice is preferred by consumers (Choudhury *et al.*, 2001). The quality of the fragrant rice is under the influence of various factors such as climate conditions, cultivation site, and genetic activities after harvest (Champagne, 2008). Post-harvest activities affecting the quality of rice include storage conditions and duration, drying method, enrichment process, and packaging. Therefore, the storage conditions such as temperature, time, and humidity have to be controlled to maintain the quality of rice (Wongpornchai *et al.*, 2004). Incorrect labelling, quality control, sorting, and adulteration are among the major issues in the rice industry. That's why rice industry utilizes standard sorting schemes based on the market criteria to determine the grains. A panel of human experts is the most common method for the evaluation of fragrant rice which detects it based on its fragrant (Rasooli Sharabiani and Khorramifar, 2022). This method,

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however, has some drawbacks such as education which may take several years, or the fatigue of the panel in the case of large number of samples which may decline the accuracy of the results (Pearce *et al.*, 2006; Lamidi *et al.*, 2023). The quality of the fragrant rice can be assessed by GC-MS which is detailed and costly (Aghili *et al.*, 2022).

Several stages of inspection may occur during transport (to determine the type and quality of the grain). Therefore, the investigation of the purity of the varieties is harder than other factors such as taste, fragrance, size, cleanness, and color. Thus, a fast, portable, and nondestructive method for rice evaluation is highly required (Abdullah *et al.*, 2016; Rasooli Sharabiani *et al.*, 2023).

Conventional quality assessment methods are mostly destructive and inefficient. The quality assessment system should be fast, precise, and low-cost. Such goals can be achieved with the help of NIR spectroscopic techniques, as they do not require sample preparation and have advantages such as non-destructiveness, fast speed, accuracy, low-cost, and pollution-free process (Sun *et al.*, 2009; Alimohammadi *et al.*, 2022; Kisalaei *et al.*, 2022). This technique is based on the radiation absorption in the near-infrared region of the electromagnetic spectrum and has been used to control the quality of agricultural and food products (Quiñones, 2018; Hu *et al.*, 2019; Williams *et al.*, 2019).

NIR spectroscopy was first used in 1968 to analyze the composition of cereals (Ben-Gera and Norris, 1968). A study used near-infrared reflectance spectrometry to investigate the quality characteristics of tomatoes such as firmness, SSC, and pH which managed to predict these characteristics non-destructively with a high correlation coefficient. The correlation coefficient for predicting SSC was 0.89 with an error rate of 0.377° Brix (Shao *et al.*, 2007).

A multispectral imaging system was used to detect adulteration in Thai jasmine rice along with chemical methods including principal component analysis (PCA), partial least squares (PLS), LS-SVM, and back-propagation neural network (BPNN). The researchers stated that four types of rice samples can be easily classified with an accuracy of up to 92% by the BPNN model. Using multi-spectral imaging technology with chemical methods, rapid and non-destructive identification of the authenticity of Thai jasmine rice can be performed (Liu *et al.*, 2021).

In another study (Fu *et al.*, 2017), NIR was used together with the in-combination one versus one least square support vector machine (IC-OVO-LS-SVM) to classify Chinese Ganoderma lucidum. The results showed that NIR and machine learning algorithms can be employed for identification and classification in the food industry. FTIR is among other non-destructive methods, which uses interference between two IR beams to produce a signal called interferometer. FTIR spectroscopy has long been used to detect food adulteration and authenticity in virgin oil (Rohman and Man, 2010; Valand *et al.*, 2020), honey (Rios-Corripio *et al.*, 2011; Gok *et al.*, 2015) and beef (Alamprese *et al.*, 2013; Nunes *et al.*, 2016). It has also been utilized to detect adulteration and authenticity in spices and plants such as saffron and oregano (Black *et al.*, 2016; Lohumi *et al.*, 2017; Petrakis and Polissiou, 2017; Wielogorska *et al.*, 2018) and nectar and jam (Miaw *et al.*, 2018). The reason for the increase in the use of FTIR can be related to the decrease in the cost of the analyses, the increase in the resolution of the analyses, as well as the rise in the demand for a lower instrument limitation for detection (Hoffman, 2013). Although FTIR spectroscopy is a low-cost and rapid screening technique in many food products, one of its drawbacks is that information about individual compounds or components in a complex mixture cannot be extracted and must be supplemented with other techniques (Valand *et al.*, 2020). Several studies have been conducted on combining spectroscopic techniques to improve prediction models (Bevilacqua *et al.*, 2012; Lee *et al.*, 2015; Ordoudi *et al.*, 2017; Quack and Merkt, 2011).

The objective of this research was to assess the capability and precision of Vis/NIR and FTIR spectroscopy in identifying pure rice varieties, specifically Hashemi rice, in comparison to four impure rice varieties. The study aimed to determine whether these spectroscopic techniques could effectively distinguish between the authentic rice variety and the adulterated ones. By evaluating the ability and accuracy of Vis/NIR and FTIR spectroscopy, this research aimed to provide valuable insights into the potential applications of these analytical methods for quality control and authentication of rice products

2. MATERIALS AND METHODS

2.1. Preparation of samples

First, 5 varieties of rice were prepared from the Rice Research Center of Iran located in Rasht city. These 5 varieties included 1 original and high-quality rice variety named Hashemi and 4 low-quality rice varieties named Fajar, Gilaneh, Khazar, and Shiroudi. Hashemi rice itself is divided into two groups, hot and cold whose appearances are not different. The cold type has better quality in terms of cooking and taste (Rasooli Sharabiani and Khorramifar, 2022). The cold type was chosen in this research. Therefore, in the experiments, one genuine rice variety (Cold-type Hashemi) and 12 non-genuine or fake varieties (a mixture of Fajr, Gilaneh, Khazar, and Shiroudi varieties mixed with Hashemi varieties with three different percentages) were prepared. The fake varieties include 90, 80, and 70% of the Hashemi variety, in addition to 10, 20, and 30% of the inferior variety. Each data collection included 25 repetitions.

2.2. Data collection with Vis/NIR spectrometer

Vis/NIR spectroscopy was carried out using a spectroradiometer model PS-100 (Apogee Instruments, INC. Logan, Utah, USA) with a CCD detector of 2048 pixels and a resolution of 1 nm and a halogen-tungsten light source in the wavelength range of 350-1 100 nm (Rasooli Sharabiani and Khorramifar, 2022). A Reflectance standard was also used for calibration.

For each sample, spectroscopy was performed with Spectra-Wiz Spectrometer OS v5.33 (c) 2014 software, and the obtained data were recorded after averaging. This software directly extracts absorption data with no need for data convergence (Khorramifar *et al.*, 2022a and 2023a; Rasooli Sharabiani *et al.*, 2022; Tarighi and Khorramifar, 2023). Figure 1 shows the graph obtained from Vis/NIR spectroscopy for the pure (Hashmi) and poor-quality varieties after noise removal.

2.3. Data collection with FTIR spectrometer

In this study, a FTIR spectrometer with a TGS (Triglycine sulfate) detector (JASCO 4700, JAPAN) was used to record the spectra. Using this device, the FTIR spectra of the samples in the 350-7 800 cm^{-1} wave number region can be obtained with a 0.4 cm^{-1} resolution.

FTIR instruments use interference between two IR beams to produce an interferometer signal, a function of the change in path length between the two beams. This is usually achieved using a Michelson interferometer. The Michelson interferometer consists of a light source such as a mercury arc, tungsten or a spherical lamp, a semi-reflective beam splitter, and two perpendicular mirrors, one fixed and the other moving (Stuart, 2004). The limiting factor for achieving higher resolution with modern FTIR instruments is the accuracy of the optical and motion mechanisms for the moving mirrors in the interferometer. High resolution is desirable, therefore, FTIR is often today the method of choice to study and investigate food adulteration and authenticity (Valand *et al.*, 2020). The FTIR graph of different varieties (Hashemi + mixed and impure varieties) can be found in Fig. 2.

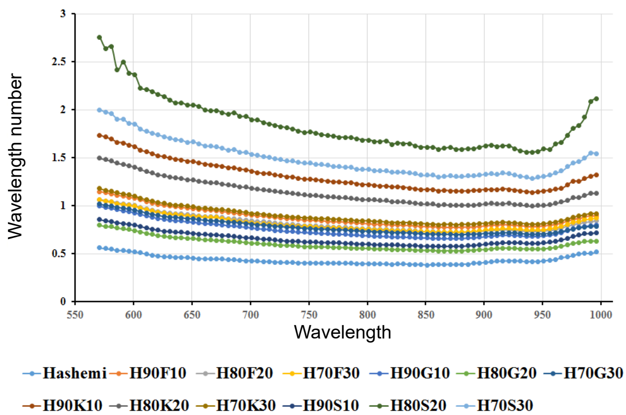

Fig. 1. Vis/NIR spectrum for different rice cultivars.

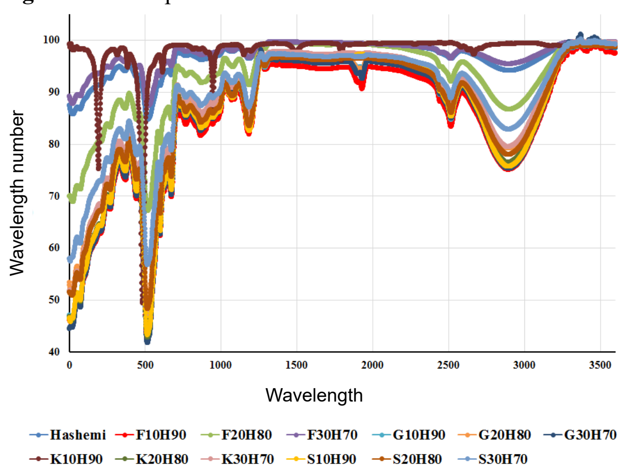

Fig. 2. FTIR spectrum for different rice varieties.

2.4. Chemometrics analysis and neural networks

Principal component analysis (PCA) is one of the simplest multivariate methods and an unsupervised technique for data clustering (Karami *et al.*, 2020a; Karami *et al.*, 2020b; Rusinek *et al.,* 2024). It is usually used to reduce the dimensionality of the data, where the best results are obtained when the data are highly correlated, positively or negatively (Abdullah *et al.*, 2015). PCA reduces the volume of multidimensional data with no loss of important information (Jolliffe, 2002; Karami *et al.*, 2020c). PCA aims to visualize the variation in a data set (such as IR spectrum) with the fewest variables (Currell, 2015), and use the linear combination of the original variables to generate new variables (Callao and Ruisánchez, 2018). This method is usually applied to distinguish adulterated and pure foods (Valand *et al.*, 2020).

Linear discriminant analysis (LDA) is the most common technique for classifying samples into predetermined classes (Berrueta *et al.*, 2007; Khorramifar *et al.*, 2023a). LDA selects independent data variables to distinguish the sample that is expected to follow a normal distribution. This method is based on linear classification functions in which the inter-group variance is maximized while the intra-group variance is minimized. This method can classify two or more groups of samples (Khorramifar *et al.*, 2021). The LDA technique is one of the techniques that require at least two defined classes and classify unknown samples based on a set of features into the class with the highest similarity (Callao and Ruisánchez, 2018). Quadratic discriminant analysis (QDA) is a variant of LDA which allows for non-linear separation of data. QDA is more flexible than LDA as it does not assume that the covariance matrix of the classes is the same. In multivariate discriminant analysis (MDA), each class is assumed to be a Gaussian mixture of subclasses (Khorramifar *et al.*, 2023a).

SVM is a supervised learning and classification algorithm based on statistical theory (Khorramifar *et al.*, 2022c). An SVM can nonlinearly map input data that are not linearly separable in a low-dimensional space to a high-dimensional space using a kernel function. A hyperplane is constructed in the high-dimensional space to maximize the distance between two classes and classify the data in the high-dimensional space. As SVM minimizes structural risks, it is considered a good classifier for non-linear data and small sample sizes (Khorramifar *et al.*, 2022b).

Decision tree organization rejects one class in each layer. The last remaining class at the bottom of the tree is considered the assigned class. The output branches of each node correspond to the possible outcome of the experiment at that node (Zareiforoush *et al.*, 2016). In this study, 3 decision tree algorithms were employed for identification and classification. These algorithms include y J48 (C4.5 decision tree learner), REP (reduced-error pruning), and LMT (logistic model trees).

Bayesian networks are models that represent a set of random variables and their conditional dependencies by a directed non-cyclic graph. Each node in the graph represents a random variable. Each random variable has a mutually unique set of possible values. One of the possible values is the true value, but we are not sure which one it is (Neapolitan, 2004). One of the main features of Bayesian networks is that they provide a suitable mathematical structure for modelling complex relationships between random variables while maintaining a relatively simple representation of these relationships (Zareiforoush *et al.*, 2016).

Multilayer Perceptron (MLP) is one of the most common types of artificial neural networks for classification. MLPs include three main layers (input, hidden, and output layers) and the layers belong to the class of feedforward networks (Haghbin *et al.*, 2023; Nayak *et al.*, 2023). That is, information passes through network nodes only in the forward direction. To classify genuine and fake rice samples, the MLP model was trained using the back propagation algorithm. This algorithm calculates the weight of the activation function for each neuron (Karray and De Silva, 2004). Various methods have been developed for error minimization in feedforward networks including gradient descent, gradient descent with momentum, Levenberg-Marquardt, and conjugate gradient (Omid *et al.*, 2010). In this research, the descent slope with the momentum approach was used to minimize the error with the momentum factor of 0.2.

In general, 60% of the data were used for training while 40% of them were applied for testing and validation.

All chemometrics and neural network analyses were performed with Unscrambler X 10.4 and weka-3-8-6 software.

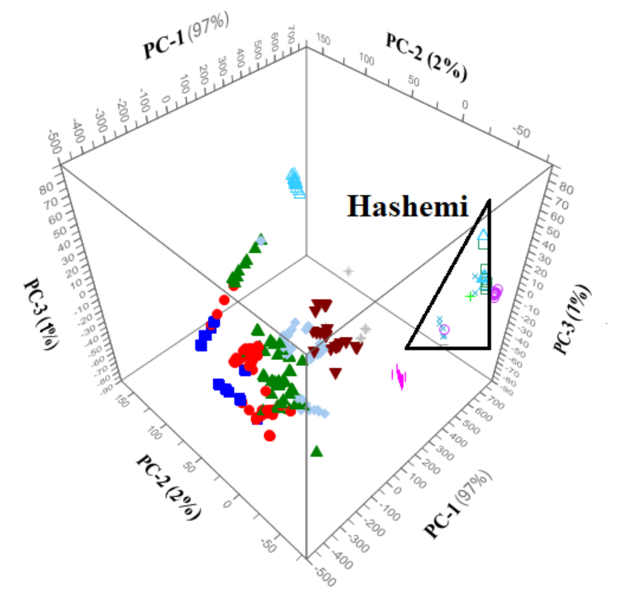

Fig. 3. PCA diagram to identify the original variety of rice with FTIR device.

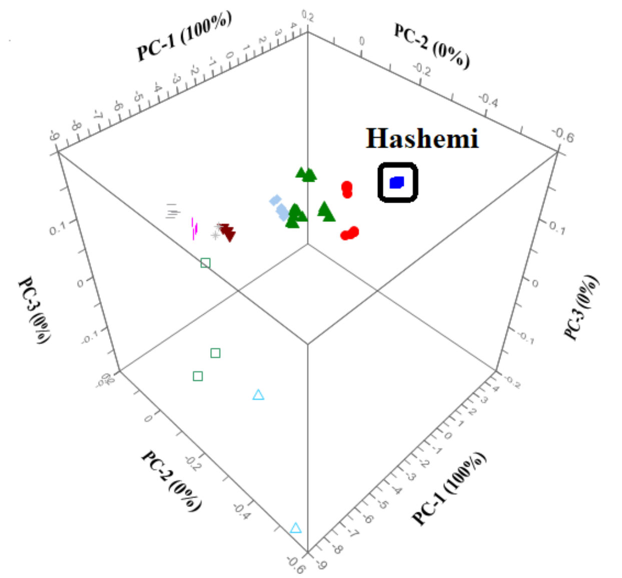

Fig. 4. PCA diagram for the detection of genuine rice variety with Vis/NIR device.

3. RESULTS AND DISCUSSION

The scores diagram (Figs 3 and 4) shows the variance of the total data equal to PC-1 (97%) and PC-2 (2%) for FTIR and PC-1 (100%) and PC-2 (0%) for Vis/NIR. The first two principal components constitute 99 and 100% of the total variance of the normalized data. For total variance higher than 90%, the first two PCs can sufficiently explain the total variance of the dataset (Khorramifar *et al.*, 2021). These results are consistent with the reports of Xu *et al.*, (2014) who expressed a PCA accuracy of 99.5% for classifying 6 rice cultivars (Xu *et al.*, 2014).

DA, SVM, and decision tree methods were also utilized to identify and distinguish original and fake cultivars based on the Vis/NIR and FTIR spectroscopic data. The LDA method can optimize the inter-class resolution according to the explanations provided in the materials and methods section. Therefore, this method was used to identify 13 rice varieties (1 original and high-quality variety plus 12 adulterated varieties) based on the data obtained from Vis/NIR and FTIR spectroscopy. In this model, the wave number of the wavelengths of each variety was considered as input (3 631 wavelengths for FTIR and 86 wavelengths for Vis/NIR) and its output included the type of rice variety (13 classes). The accuracy of distinguishing figures was obtained according to Table 1.

According to Table 1, QDA and MDA methods with Vis/NIR spectroscopy offered the highest accuracy (Figs 5 and 6).

According to Table 1, the accuracy of the DA method with the Vis/NIR spectrometer is much higher than that of the FTIR spectrometer.

Here, C-SVM and Nu-SVM methods were employed to classify 13 rice varieties. Four radial, sigmoid, polynomial, and linear basis kernel functions were used as SVM kernel functions. Similar to the LDA model, the number of wavelengths of each variety was considered as input (3 631 wavelengths for FTIR and 86 wavelengths for Vis/NIR) while the output included the type of rice variety (13 classes). The results of the SVM method are summarized in Tables 2 and 3 for FTIR and Vis/NIR spectroscopy, respectively. The highest accuracy was related to the FTIR spectrometer and C-SVM methods (with Linear and Polynomial kernel functions) with 100% training accuracy and 99.38 and 99.08% validation accuracy as can be seen in Fig. 6. Moreover, the Sigmoid kernel function had the lowest accuracy (for training and validation).

Table 1. The results of the DA method for detecting genuine rice with spectroscopic devices

Method	Accuracy (%)	
	FTIR	Vis/NIR
LDA	78.15	92.31
QDA	86.77	100
MDA	78.77	100

Table 2. Accuracy of results and comparison of Nu-SVM and C-SVM statistical models subjected to kernel functions for FTIR and Vis/NIR

Kernel function	C-SVM (%)		Nu-SVM (%)		Device
	Training	Validation	Training	Validation	
Linear	100	99.38	90.77	89.85	FTIR
Polynomial	100	99.08	86.15	85.23	
Radial	95.08	91.38	92.62	90.46	
Sigmoid	7.69	7.69	7.69	7.69	
Linear	90.77	89.23	95.38	93.85	Vis/NIR
Polynomial	90.77	86.15	95.38	89.23	
Radial	90.77	89.23	95.38	90.77	
Sigmoid	0	0	92.31	87.69	

Table 3. Results and comparison of decision tree, MLP, and Bayesian models for Vis/NIR and FTIR

Algorithm	FTIR		Vis/NIR	
	Accuracy (%)	RMSE	Accuracy (%)	RMSE
MLP	92.31	0.0967	96.93	0.0810
Bayesian	92	0.1040	100	0
J48	92.31	0.1065	100	0
LMT	98.77	0.0428	100	0.0752
REP	90.46	0.1166	99.69	0.0191

According to the results listed in Table 2, the accuracy of SVM method with FTIR spectrometer was higher than the Vis/NIR (in contrast with DA method). The results obtained in DA method are completely the opposite (*i.e.* the precision of DA was higher in NIR rather than FTIR). This difference can be assigned to the differences in the wavelength ranges of the two mentioned devices. The wavelength range of FTIR is higher than NIR and even some changes can be observed in higher wavelengths (above 2 500 nm) which are not experienced in NIR (due to the lower wavelength range).

The Vis/NIR method achieved the rice classification accuracy of about 90% which is in line with the results of Kaur and Singh (2013) who performed rice classification with a vision machine and reported the accuracy of 90% (Kaur and Singh, 2013). It is also clear that the accuracy of our research with the FTIR spectrometer is higher than the results of this research.

Table 3 lists the accuracy of the results of MLP, Bayesian, and 3 different decision tree algorithms for both spectrometers.

According to the table, it is quite obvious that the accuracy of MLP, Bayesian, and decision tree networks was higher with Vis/NIR spectroscopy rather than FTIR. Furthermore, the decision tree algorithm offered very high precision. Zareiforoush *et al*. (2016) utilized computer vision and MLP, Bayesian, and decision tree algorithms for the qualitative classification of milled rice grains (Zareiforoush *et al.*, 2016) and reported respective classification accuracies of 96.67, 97.51, and 97.98 which are similar to the present research.

Abdullah *et al*. (2015, 2016) identified and classified 17 rice samples into 4 categories using an olfactory machine. According to their report, the accuracy of the SVM method was equal to 100% which was the best classification method for rice varieties. They also expressed that the KNN method is the second best approach for classifying rice samples (Abdullah *et al.*, 2016). Their results were in line with the findings of the current research.

Moreover, Jana *et al*. (2011) used E-nose along with ANN, PCA and LDA techniques to distinguish fragrant rice from non-fragrant one with respective accuracies of 93, 96.5, and 80%. Meanwhile, the accuracy of LDA method was the same as the accuracy of LDA in our study.

In a study which employed a machine vision and MLP method to identify rice cultivars, the accuracy of 84.83% was reported (Gujjar and Siddappa, 2013), which is lower than the results of the present research.

The results of the PCA method in the work of Han *et al*. (2016) to detect adulteration in oil were highly consistent with the outcomes of the PCA method in our research. They identified adulteration in oil with hyperspectral imaging and reported the accuracy of 99% for PCA method under UV illumination in the IR spectrum.

In another study, Liu *et al*. (2017) utilized multispectral imaging and chemometrics methods to detect sucrose adulteration in tomato paste. They reported that the accuracy of PLS and BPNN methods was 93% lower than the other two meth-

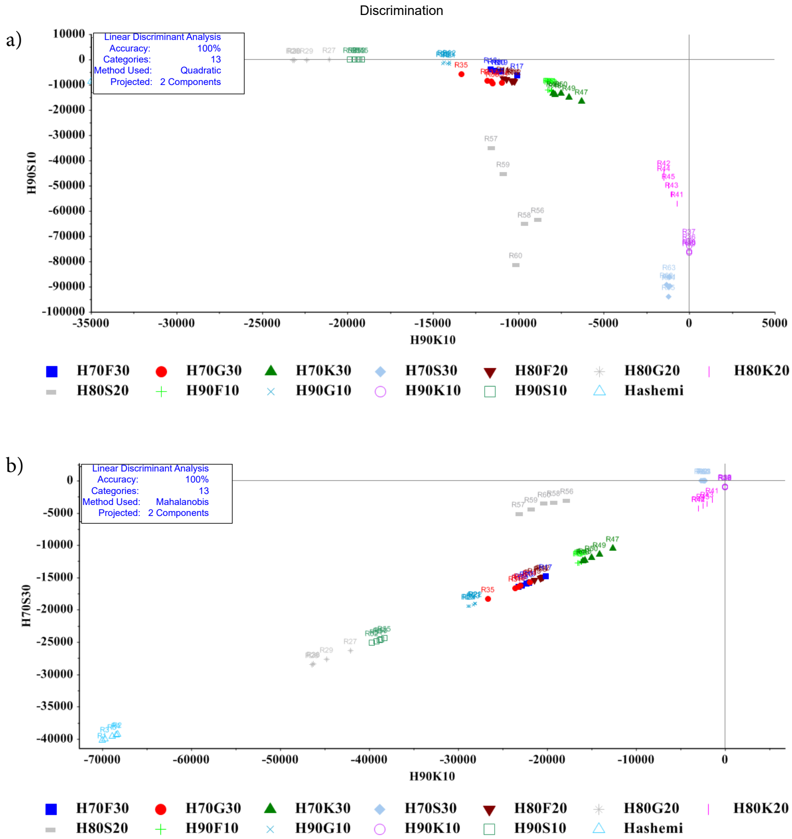

Fig. 5. Results of DA analysis for NIR device: a) QDA, b) MDA.

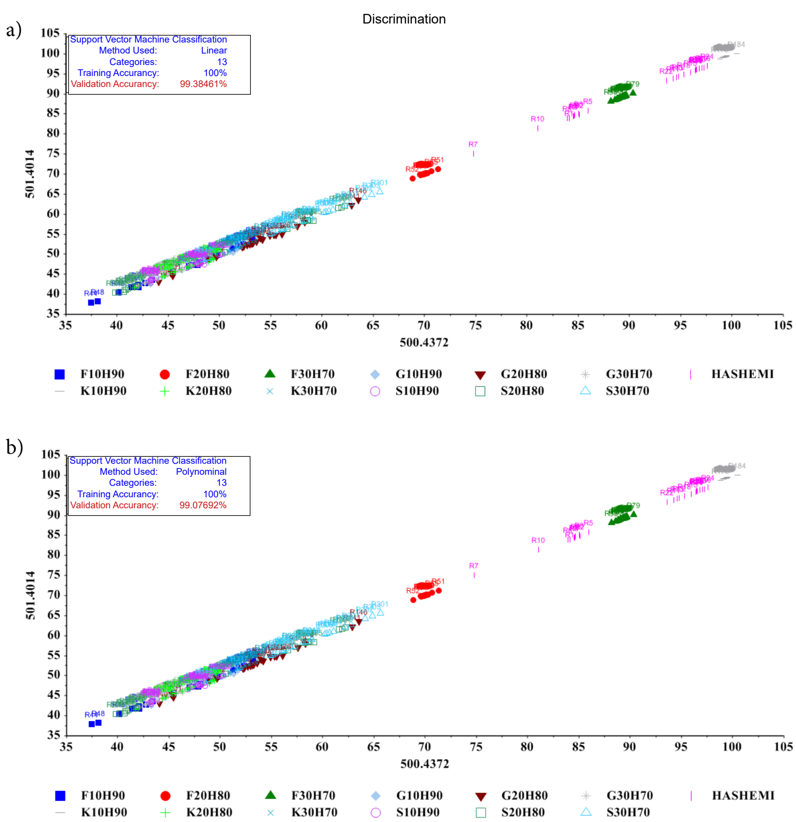

Fig. 6. Results of SVM analysis for FTIR device: a) linear, b) polynomial function.

ods (LS-SVM and PCA), while the accuracy of LS-SVM and PCA methods was 96 and 98%, respectively. Therefore, the PCA method had a good response compared to other methods.

Georgouli *et al*. (2017) used FTIR and Raman spectroscopy methods with different methods to detect adulteration in olive oil. They found similar accuracy of FTIR and Raman detection (Georgouli *et al.*, 2017).

Comparing the results of two spectrometers, the accuracy of Vis/NIR was higher than FTIR which is in line with the reports of Bevilacqua *et al*. (2012) who reported higher precision of Vis/NIR spectrometer in prediction of adulteration in onion and olive powder when compared to FTIR spectrometer (Bevilacqua *et al.*, 2012).

Furthermore, since the fluorescence signal occurs at sub-excitation energies, it often interferes with sample analysis; thus, the sensitivity of Raman spectroscopy is generally lower than Vis/NIR and FTIR (Smith and Dent, 2019).

4. CONCLUSIONS

According to the results of this research, visible–near–infrared and Fourier-transform infrared spectrometers can generally identify genuine (Hashemi) and adulterated rice (Hashemi mixed with other varieties) at high accuracy. All the methods in this research were highly accurate, except for the support vector machine method with the Sigmoid function, which exhibited high error rate.

Other spectroscopic methods are recommended to be used to identify the best method for detecting the authenticity and adulteration of Hashemi rice. One of the examples is the Raman spectroscopy whose one of the advantages compared to visible–near–infrared and Fourier-transform infrared spectrometers is that it does not show water interference, which could otherwise be problematic when analyzing foods without prior sample preparation. On the other hand, the major disadvantage of Raman spectroscopy is the reliance on its detection of a weak fluorescence signal, hence, requiring a powerful and expensive excitation source.

Conflicts of Interest: The authors declare no conflict of interest.

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