PhD THESIS
– ABSTRACT –

Optical sensors and advanced methods with application on food additives and beverages analysis

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1. Motivation

Over the years the habits of the people, regarding the procuring, preparation and storage of food products have changed [1]. Centuries back, people procured and prepared food by themselves. Man used to hunt, whereas women cooked the meat and other plants that grew around the household for the entire family. Old habits started to slowly change, animals were tamed and domesticated, fruits and vegetables were planted around the households [1]. Centuries passed and people continued growing fruits and breeding animals for their families, for sale or for barter.

People's habits regarding food acquisition and preparation started to change mostly in the XX century when technological advances allowed farmers to use horse-drawn combine and, later on, steam powered tractors. Slowly, mechanized agriculture replaced the old ways of agriculture. In addition, technological progress determined the apparition of the first factories that prepared food for sale. The products were placed in packages and sent to selling points where people could buy them according to their preferences, needs and budget.

Nowadays, a lot of products are available at markets and supermarkets. The procurement of food has become a lot easier, but it has also come with some disadvantages. Firstly, people are no longer directly involved in the preparation of the food products that they consume. Therefore, they cannot control or change the ingredients that are used in foods or beverages. The downsides may concern the quality of the ingredients or the replacement of some ingredients with their chemical and cheapest version. For example, vanillin is a natural substance found within vanilla beans, but it can also be chemically synthesized with reduced costs [2]. Also, sugar can be replaced by chemically synthesized sweeteners, known as artificial sweeteners, which are more sweet than regular sugar [3].

Artificial sweeteners are a type of food additive. Additives can be classified in several categories based on the main functions. For example, there are additives that counteract the spreading of bacteria, others determine a texture (creamy, crispy), control the colouring or regulate the acidity of the products. All the additives are available on the Codex Alimentarius provided by FAO (Food and Agriculture Organization) and WHO (World Health Organization) [4], [5]. Each additive was associated with an E number. In a single product may be one or more food additives. Table 1.1 presents additives within four different classes, their acceptable daily intake (ADI), the molecular mass, the E number and the molecular formula.

Table 1.1 Food additives (adapted from [6])

Food additive	Molecular Formula	E number	Molar mass [$g \cdot mol^{-1}$]	Type of additive	ADI [mg/kg bw] [7]
Potassium sorbate	$C_6H_7KO_2$	E202	150.218	Preservative	0-25
Sodium benzoate	$C_7H_5NaO_2$	E211	144.105	Preservative	0-5
Ascorbic acid	$C6H8O6$	E300	176.12	Antioxidant	Not specified
Aspartame	$C_{14}H_{18}N_2O_5$	E951	294.307	Sweetener	0-40
Saccharin	$C_7H_5NO_3S$	E954	183.18	Sweetener	0-5
Acesulfame potassium	$C_4H_4KNO_4S$	E950	201.242	Sweetener	0-15
Citric acid	$C6H8O7$	E330	210.14	Acidity regulator	Unlimited
Trisodium citrate	$Na_3C_6H_5O_7$	E331	294.10	Acidity regulator	Unspecified

The possible effects of these additives have been intensively studied (aspartame [8]– [11], acesulfame potassium [12], saccharine [13]). The health effects that might be induced by these additives and the need of people to avoid certain ingredients due to health-related

problems (diabetes, lactose intolerance, allergies) represent valid motives for finding precise and fast analysis methods for food products and beverages. Among the analysis techniques that were found in literature can be enlisted spectroscopy, colorimetry, interferometry and HPLC (high-performance liquid chromatography). What these techniques have in common is the use of light for the analysis of the samples. For example, UV (ultraviolet) spectroscopy was used for the differentiation and classification of wines samples [14], fluorescence spectroscopy for honey analysis [15], whereas FTIR (Fourier-transform infrared spectroscopy) analysis was used for contaminated milk discrimination [16].

The study of light sciences began after the mathematical model of the electromagnetic field was developed by the scientist James Maxwell. From that moment on, light was not considered only a ray, but was also defined as a wave. Light was also of great interest for scientists such as Isaac Newton, Christian Huygens or Thomas Young. Up until this moment, light has been used in various domains: medicine [17], military [18], astronomy [19], biology [20], not only for the analysis of edibles [15], [21]– [24].

This thesis, entitled "Optical sensors and advanced methods with application on food additives and beverages analysis", presents a short theoretical study of optical sensors and optical analysis methods, study that is followed by the presentation of the research results that were obtained. The research focused its interest on the analysis of beverages and food additives utilizing optical sensors and techniques. Regarding, the large number of research papers that target the possible effects of food additives, the increased variety of food products and beverages that may contain these substances, and the diversity of the techniques that are used for the analysis, it can be concluded that the discrimination and characterization of food additives is an up-to-date research topic of interest great interest for the modern society.

2. Objectives

This work proposes a theoretical study on the types of optical sensors and advanced analysis methods based on light. The main objective of the paper regarded the use of the studied sensors and optical methods for the analysis of food additives and beverages. Therefore, the main objective was divided into four objectives as follows:

- The testing of experimental setups constructed with low-cost optical devices for the analysis of milk and juice samples.
- The analysis of food additives using ultraviolet spectroscopy and the spectra classification using machine learning techniques.
- The classification of the UV spectra of simple and mixed solutions of food additives using artificial and convolutional neural networks (ANN and CNN).
- The testing of holographic sensors recorded on a commercial photopolymer with various food additives, the recording of the optical displacement caused by the analytes and the determination of the relationship between the concentration and the optical response of the sensors.

3. Methodology

A research methodology was established to achieve the proposed objectives. The methodology included three main stages: documentation and theoretical study, research and dissemination.

3.1 Documentation and theoretical study

In the first phase, a documentation was carried out in the field of optical sensors and advanced analysis methods, which allowed to create an overview image on the subject. Various types of optical sensors were identified, and real applications were carefully analyzed. The functioning principle, the advantages and the disadvantages of each technique were compared. The process of study and documentation was carried out continuously during the PhD studies in order to be familiar with the main research topics of other teams and discover new niches that could be treated, studied and researched.

3.2 Research

The theoretical study was followed by research work that was conducted in collaboration with various research groups. The research stage began by testing experimental setups with low-cost optical devices such as LEDs (light emitting diodes), optical sensors and non-specialized spectrometers. The experimental setups were used to analyze the time evolution of non-alcoholic beverages such as juices and milk. This project was accomplished with the help of the "Optoelectronics" group at the Electronics, Telecommunication and Information Technology Faculty from the Technical University of Cluj-Napoca.

The research work was continued at the Technical University of Riga, in the „Biomedical Engineering and Nanotechnologies" (BINI) department. The mobility was possible through an ERASMUS scholarship. During this mobility the theoretical basis of optically induced electrophoresis, and microfluidics were studied. A tri-dimensional model for the coupling of a LASER (light amplification by stimulated emission of light) with an optical fiber was designed, 3D-printed and assembled. The device was built for the light analysis of cells.

Another scholarship was obtained in the project "Network of excellence in applied research and innovation for doctoral and postdoctoral programs/InoHubDoc". The scholarship facilitated the collaboration with the Department of Polymer Composites from the Raluca Ripan Institute for Research in Chemistry from Cluj-Napoca. During this collaboration, the work focused on the analysis of food additives using UV spectroscopy followed by spectra classification using artificial intelligence techniques.

Finally, holographic sensors were studied during an ERASMUS mobility at the University of Zaragoza. The mobility started with a theoretical study on holography and it was continued with the testing of the holographic sensors recorded on a commercial photopolymer with various substances. The study of holographic sensors was only possible with the help and guidance of the "Laboratorio de Holografía" members within the "Tecnología Óptica Láser" group (Applied Physics Department, University of Zaragoza).

The research work was consolidated by participating in three summer schools and one young research conference. One of the summer schools is called "V International School on Light Sciences and Technologies V-ISLiST" and focuses on light sciences. It took place in Santander, Spain in June 2022.

The other two summer schools, called ELaRa, focused on pedagogical subjects and took place in September 2022 and June 2023. Also, the participation in the young research conference called "1ª Jornada de Jóvenes Investigadores del Grupo TOL – 1ª JOJITOL", organized at the University of Zaragoza, was an interesting experience which provided great knowledge regarding the research topics of other young scientists.

3.3 Dissemination

The results obtained during the entire PhD study were disseminated at four international conferences and by publishing an article in the journal „Sensors" (Q2). Another article is currently under review at the journal "Optics and Laser Technology".

Furthermore, the results were also appreciated in a Novice Insights organized by the Electronics, Telecommunication and Information Technology Faculty from the Technical University of Cluj-Napoca. The paper was awarded the second prize at the Master/PhD section in 2023.

4. Thesis Structure

This thesis was divided into two main parts: "Current Stage of Knowledge" and "Personal Contributions". The first part encompasses chapters 1 and 2, whereas the second part includes chapters 3-8.

4.1 Current Stage of Knowledge

The first part of the thesis includes a theoretical study of advanced methods used for sample analysis and for data processing. This part was divided into two chapters: "Optical sensors and analysis techniques" and "Machine learning and deep learning techniques".

The first chapter encompasses three subchapters: "Optical sensors", "Optical analysis techniques" and "The analysis of food additives and beverages". This chapter offers a short presentation regarding the types of optical sensors and analysis techniques, ending with the presentation research studies where beverages and food additives were analyzed using the aforementioned techniques or sensors.

The second chapter was also partitioned into three subchapters: "Artificial Neural Networks", "Convolutional Neural Networks" and "Applications". In these subchapters, the architecture and the parameters of artificial and convolutional neural networks are presented and explained. The third subchapter focuses on real applications where optical techniques were used with machine and deep learning technique.

4.1.1 Optical sensors and analysis techniques

Chapter 1, "Optical sensors and analysis techniques", presents an introduction regarding the types of optical sensors and the analysis techniques that imply the use of light. Optical sensors have been used in medicine for glucose measurements [25], in health monitoring devices [26], for soil analysis systems [27], but also for food analysis [28].

The theoretical study has established that optical sensors can be divided into two main categories. The first category includes sensors that can measure light intensity and transform it into an electrical signal (which can afterwards be processed within an electronic circuit) [29]. These devices are known as photodetectors and include three types of sensors: photoresistors, phototransistors and photodiodes. These sensors can be integrated into CMOS and CCD image sensors or inside appliances such as spectrometers [30] or chromatographs [31].

The second category of optical sensors includes holographic and optical fiber sensors. These devices are able to measure external parameters such as temperature, strain, humidity or the presence of a certain analyte based on the changes that appear in their optical parameters. Optical fiber sensor were used to measure temperature [32], [33] or humidity [[34]–[36], whereas holographic sensors were utilized for pH detection [37], [38] and glucose determination [39], [40].

The theoretical study was continued with the identification of analysis methods that use light for the sample characterization. Among the techniques can be listed spectroscopy, interferometry, holography and HPLC. Spectroscopy is an analysis technique which studies how much light can a sample absorb/transmit [30]. Spectroscopic methods can be classified based on the type of electromagnetic radiation that is used in the analysis. There is ultraviolet spectroscopy, infrared spectroscopy, fluorescence spectroscopy, but also FTIR spectroscopy (Fourier-transform infrared spectroscopy).

Interferometry is another analysis method that uses light waves. It is based on the interference of two light waves that are emitted by a monochromatic light source. The light waves come from the same source, but they travel on distinct paths until they meet again. An interference pattern is created when the waves meet, and depending on the application the pattern may contain information about the substance that is analyzed. The devices used for this technique are called interferometers. The most commonly used interferometers are Mach-Zehnder, Michelson, Sagnac şi Fabry-Perot.

Another technique that uses the interference of waves is holography [41]. Tri-dimensional images can be recorded with this technique. The main components that are required for recording a hologram are: a coherent and monochromatic light source (e.g. LASER), a material sensitive to light which is known as a photopolymer and the object whose image will be captured in the hologram.

HPLC is a separation technique by which chemical species within a mixture can be separated, identified and quantified. The technique requires the use of a pump, a column, injectors and a photodetector [42]. Normally, there is an array of photodetectors within the appliance which can detect radiation in the visible and ultraviolet domain.

The theoretical study has highlighted that the analysis techniques based on light are currently very frequently utilized. Recent research papers describe the analysis of food products, beverages and food additives using UV spectroscopy [43], [44], infrared spectroscopy [45], [46], holography [47] or HPLC [48]–[51].

4.1.2 Machine learning and deep learning techniques

Chapter 2 describes the operation principle of machine and deep leaning techniques. These techniques are part of artificial intelligence methods and were successfully used for voice and face recognition, email sorting or for self-driving auto vehicles.

The algorithms can learn the most important features of the datasets, make new predictions as accurate as possible and improve their performance. They must firstly be trained and tested, steps followed by the validation of the models using novel datasets. The initial dataset must be divided into three subsets: one for training (it is the largest dataset, containing approximately 80% of the data recordings), one for testing (when the predicted results are compared with the actual results) and one for validation (when the system is able to identify new datasets based on the two previous steps). Examples of machine learning techniques are decision trees, k-NN (k-nearest neighbors) or SVM (support vector machine).

Deep learning techniques include methods whose operating principle is similar to the one of the human brains. The algorithms are based on artificial neural networks that are built from several layers that contain artificial neurons. The architecture of an artificial neural network includes an input layer, one or more hidden layers and an output layer. These layers are also called fully connected layers. Beside these layers, in a convolutional neural network, are also present convolutional layers followed by pooling layers. The convolutional and pooling layers are separated from the fully connected layers by a flatten layer.

The theoretical study included articles regarding the architecture of deep learning techniques [52]–[55] and articles where machine and deep learning techniques were used together with optical analysis techniques for beverage analysis [23], food analysis [56]–[59] or food additives identification [60]. For example, improved SVM was combined with terahertz

spectroscopy for the analysis of additives containing coumarin [60] but also with laser-induced breakdown spectroscopy for the analysis gelatine samples [56].

4.2 Personal Contributions

The second part of the thesis presents the research results obtained throughout the PhD period. In the first part of the thesis, optical sensor and analysis methods were theoretically studied. In the second part, all the acquired knowledge about optical sensors and advanced techniques was used for practical applications. The experimental work focused on the analysis of liquid samples of food additives and beverages. Therefore, the second part was divided into five chapters: "Introduction and objectives", "Experimental setups for beverage analysis", "Machine learning techniques for food additives classification", "Deep learning techniques for food additive classification", "Holographic sensors" and "Final Conclusions".

Chapter 3 makes a brief introduction in the thesis thematic and presents the main objects that were to fulfill. **Chapters 4-7** detail the solutions proposed for the analysis of beverages and food additives and the research results. This part is finalized with **Chapter 8**, which is dedicated to presenting the main conclusions and highlighting the personal contributions of this thesis.

4.2.1 Optical analysis setups

Chapter 4, called „Experimental setups for beverages analysis" presents experimental setups used for analyzing milk and juice samples (natural and commercial). It was desired to use low-cost optical devices such as LEDs (light emitting diodes), optical sensors and non-specialized spectrometers in the experimental setups. At the moment, there is specialized laboratory equipment that can examine food and drink samples, the drawback regarding its price and its large dimensions. Therefore, the goal was to test if a simple experimental setup with LEDs and optical sensors could indicate a change of the analyzed samples.

This study was based on two articles [61], [62] that described analysis systems using low-cost devices. The first one presents the use of an UV LED and the ML8511 sensor for the detection of nitrites and nitrates within water samples [61]. The second article uses the TCS200 color sensor to detect the correlation between the absorbance spectra of erythrocytes and the number of cells within blood samples [62]. Other applications used UV LEDs for water sterilization [63] and infrared devices (LEDs and phototransistors) for isopropyl alcohol detection [64].
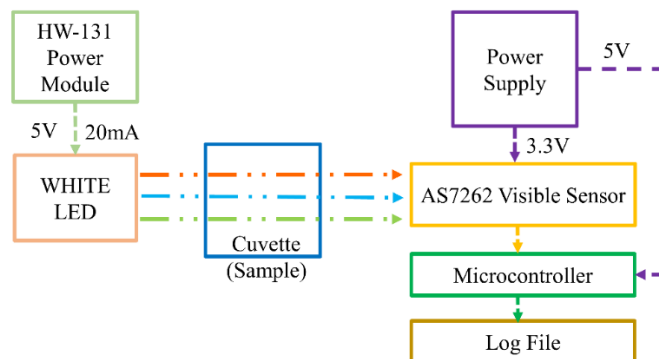

Figure 4.1 Experimental setup (adapted from [65] © 2021 IEEE)

The proposed experimental setup for juice and milk analysis was based on the following components: the AS7262 sensor with six channels in the visible domain [66] and a white LED [67]. A cuvette was placed between the light source and the sensor. The juice or milk sample

was deposited in the cuvette. The analysis relied on analyzing and comparing the values acquired by the sensor over a period. The values were expected to change due to a natural process known as fermentation. Figure 4.1 shows the experimental setup constructed with the two optical components. The LED was powered using a HW-131 module which can provide voltages of 3.3V and 5V. The sensor was connected to the microcontroller ATMEGA328P from Arduino Board. The board read the values obtained from the sensor.

The emitter, the cuvette and the sensor needed to be perfectly aligned so the emitted light would come in contact with the sample, and if not absorbed, pass through it and reach the sensor. Alignment was possible through 3D (tri-dimensional) models which were designed according to the dimensions of the components. The structures were constructed using a 3D printer. Figure 4.2 illustrates the tri-dimensional model for the sustaining structure used to hold the optical devices. The lateral opening from Figure 4.2 (a) and (b) allowed radiation to pass from the source to the sensor. Figure 4.2 (c) shows the printed structure with the optical sensor attached to it.

The optical sensor can be attached to the structure using the two lateral extremities, which were specially designed according to the board dimensions. The extremities are shown in Figure 4.2 (b). By introducing the opening of the sensor's board in the lateral extremities the sensor is kept fixed, preventing erroneous measurements. To prevent the sample from evaporating a 3D model for a lid was designed for the cuvettes. The cuvette lids, the cuvette, the sensor and the sustaining structure are shown in Figure 4.2 (c).

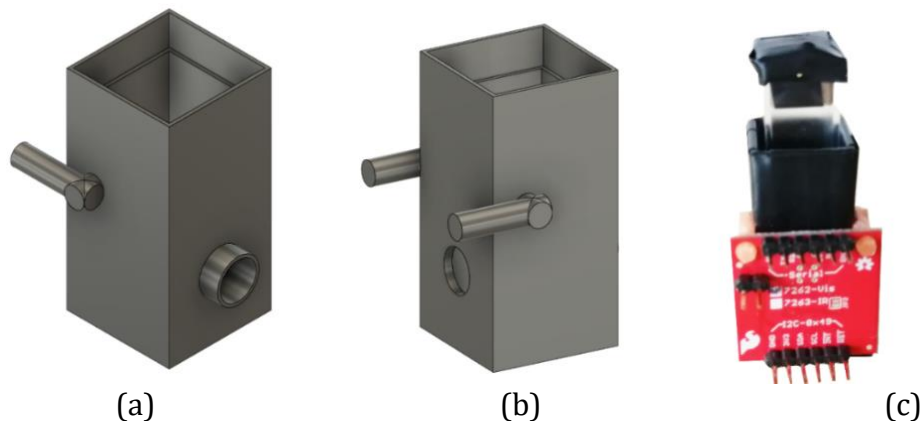| (a) | (b) | (c) |

Figure 4.2 Sustaining structures (a) left view (b) right view (c) 3D printed structure, AS7262 sensor, cuvette and cuvette lid [65] © 2021 IEEE

Three sets of measurements were made with milk samples. The results of the first set are shown in Figure 4.3 (a). The light intensity detected by the sensor increased over the first days of analysis. During the seven-day analysis the texture of the milk sample changed from liquid to solid (at the end of the measurements). Therefore, based on the results it was concluded that the rise might be an indicator of a natural process known as fermentation.

To verify if the measurements are repeatable, the experiment was repeated two more times. Figure 4.3 (b) presents the results of the second set, the rise in the measured values being also visible. Similar results were obtained for the third set of measurements which is represented in Figure 4.3 (c).

The experimental setup was also used with commercial juice samples. Three types of juices were chosen and analyzed within an eleven-day period, but there was no intensity change visible. The results for one the commercial juice are shown in Figure 4.3 (d). The study was continued with samples of natural juices. The three types of fruits that were analysed are presented in Table 4.1. Besides the intensity values read by the optical sensor, the pH was also monitored using a digital pH-meter.
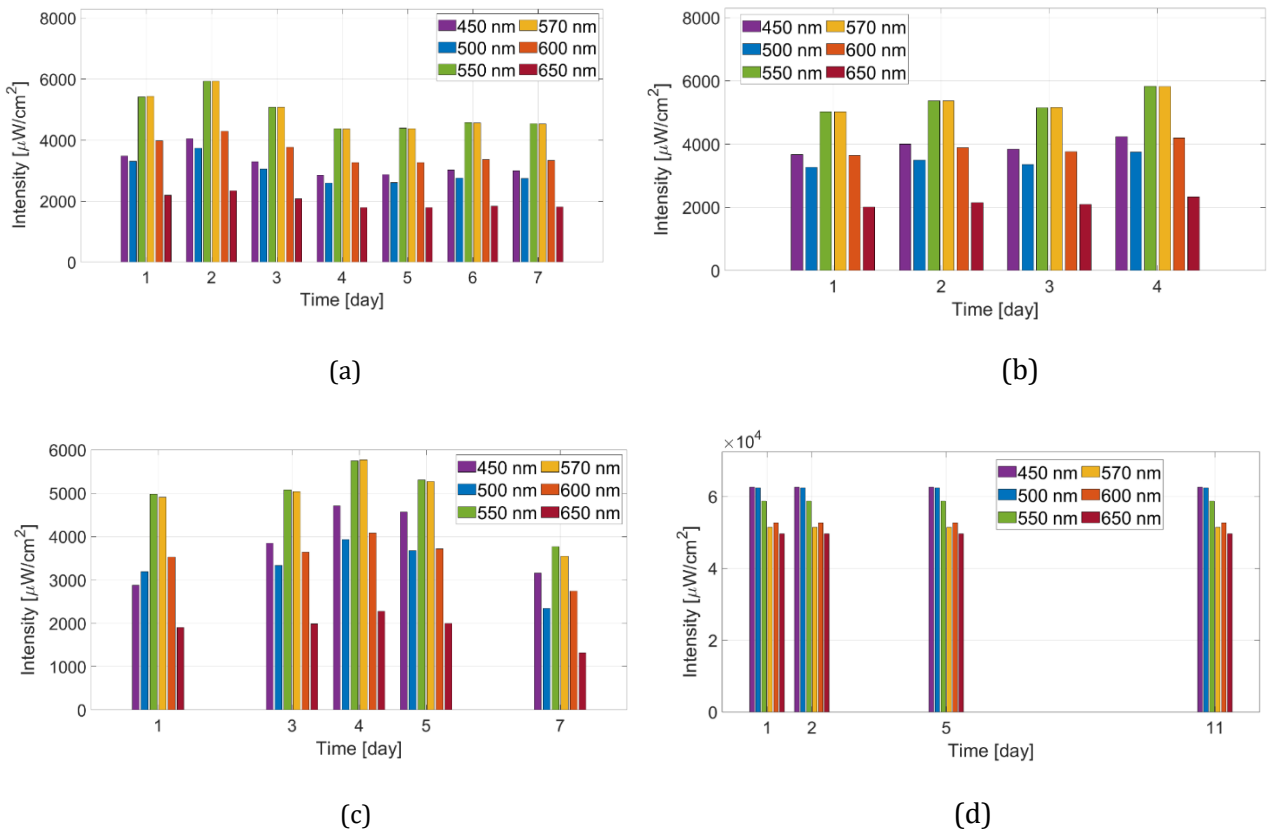
(a)

(b)

(c)

(d)

Figure 4.3 Analysis results for juice and milk samples (a) first set of measurements (milk) (adapted from [65] © 2021 IEEE) (b) second set of measurement (milk); (c) third set of measurements (milk); (d) measurements for a commercial juice sample

Figure 4.4 (a), (b) and (c) show the intensity results obtained with the AS7262 sensor for the three citrus juices, whereas Figure 4.4 (d) depicts the pH evolution of the samples. The results from Table 4.1 have indicated that the pH values decrease, whereas the light intensity read by the sensor increases, especially for the 450 nm channel (that the quantity of light absorbed by the samples decreased).

Table 4.1 pH of natural citric juices

Citrus Fruit	Reference pH [68]	Initial pH	Final pH	ΔpH
Lemon juice	2.34 ± 1.8	2.7	2.4	0.3
Orange juice	3.60 ± 1.9	4	3.6	0.4
Grapefruit juice	3.00 ± 2.5	3.5	3.3	0.2

The results showed that the experimental setup with low-cost devices may provide information regarding the decay of the analyzed samples. The results obtained with the AS7262 sensor were compared to the ones obtained in other study with the AS7265x. This device is a smart spectral sensor that can measure absorbance values for a wide domain of wavelength (410 and 940 nm), compared to the AS7262 sensor which can detect the light intensity of only six channels within the visible domain. This device was used for the analysis of milk, honey and other liquid samples [69]. Some of the milk samples were adulterated with urea to compare the absorbance values obtained for non-adulterated and adulterated samples. The results have indicated that the pure milk samples had an absorbance value higher than the ones that were adulterated. Similarly, in this study, milk samples with high intensity values (low absorbance values) were considered to be fermented.
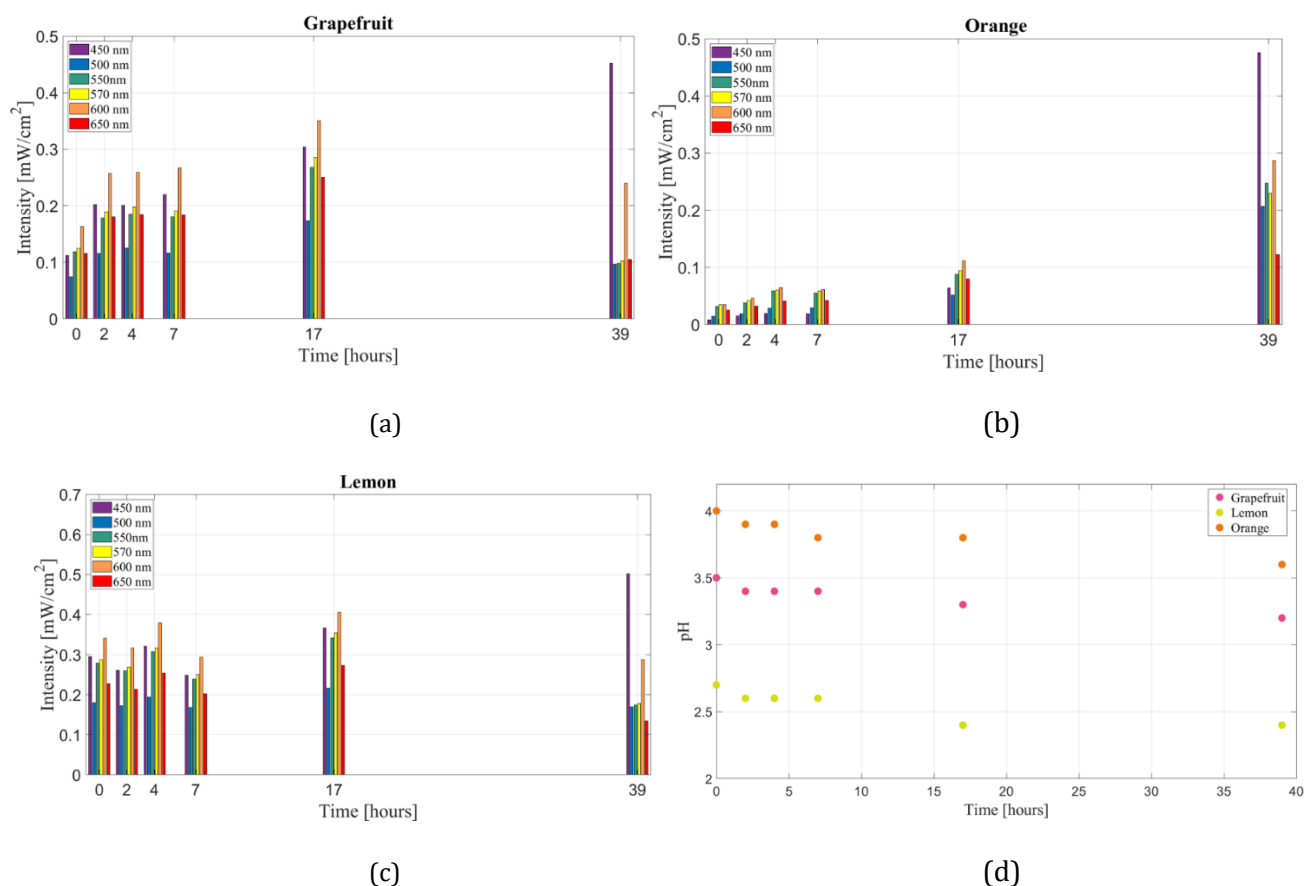
Figure 4.4 Experimental results (a) grapefruit juice; (b) orange juice; (c) lemon juice; (d) pH measurements for all juices from [70]

4.2.2 Machine learning techniques for food additive classification

In **chapter 5,** called "Machine learning techniques for food additive classification" is presented an approach for the classification of five food additives using UV spectroscopy and machine learning techniques.
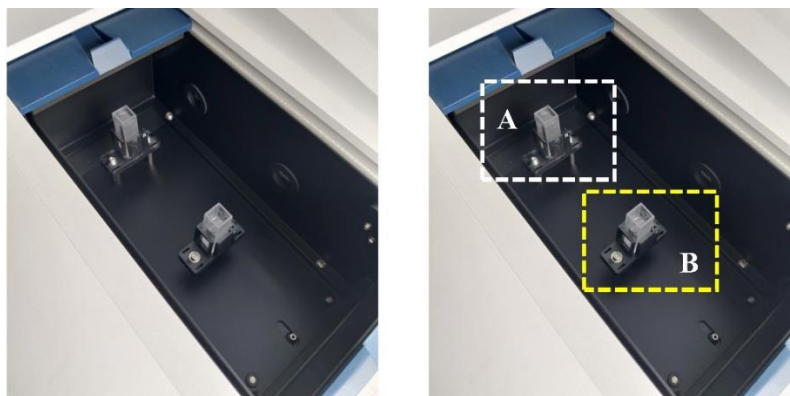

Figure 4.5 Cuvette positioning into the spectrometer (A. reference cuvette; B. sample cuvette)

The spectroscopic measurements were performed on liquid sample, the additives being dissolved in distillate water prior to the analysis. The liquid samples were placed in quartz cuvettes and positioned inside the UV spectrometer (Figure 4.5) [71]. One cuvette was used as the reference and contained the solvent in which the solute was dissolved, whereas the second was filled with a sample. The sample was prepared by dissolving the food additive into distillate

water. The acquired spectra were used for training, testing and validating the machine learning algorithms. To be able to classify the spectra, more than one spectrum of each additive was necessary. Therefore, several samples with different molarities were prepared for each additive. Table 4.2 shows the molar concentrations ranges for the five additives.

Table 4.2 Molar concentrations [6]

Food additive	Minimum molar concentration [M]	Maximum molar concentration [M]
Acesulfame potassium	0.0000937	0.000625
Aspartame	0.00175	0.01
Sodium benzoate	0.0000137	0.0005
Potassium sorbate	0.00000312	0.000195
Saccharin	0.000292	0.00225

In Figure 4.6 (a) the maximum absorbance peak has a value of 1.4 A.U (absorbance units) and corresponds to a solution with a molar concentration of 0.000156 M. The absorbance value decreased for solutions with lower molar concentrations (0.0000781 M and 0.0000390 M) confirming the Beer-Bouguer-Lambert Law. The spectra of the five additives were different by shape, number of absorbance peaks and the wavelength where the peaks were located. Therefore, the additives could be identified and classified based on the UV spectra.

The feature extraction was necessary in order to classify the spectra using machine learning algorithms. Therefore, spectral parameters were automatically calculated based on the acquired spectra. Figure 4.7 shows four of the parameters calculated using the spectra. These parameters are: FWHM (full width half maximum), λ_P (wavelength where the maximum absorbance peak appears), λ_L and λ_R (wavelength where the absorbance is half of the maximum absorbance peak). The FWHM parameter can be calculated as the difference between the wavelength for half of the maximum absorbance value). The Skewness parameter and the difference between λ_P şi λ_L, respectivelly between λ_R şi λ_C were also determined in order to establish if the spectra were symmetrical. Other parameters were Kurtosis and the relative errors of the following parameters λ_P, λ_L, λ_R şi FWHM. Table 4.3 presents some of the parameters determined for five solution different additives.

(a)   (b)
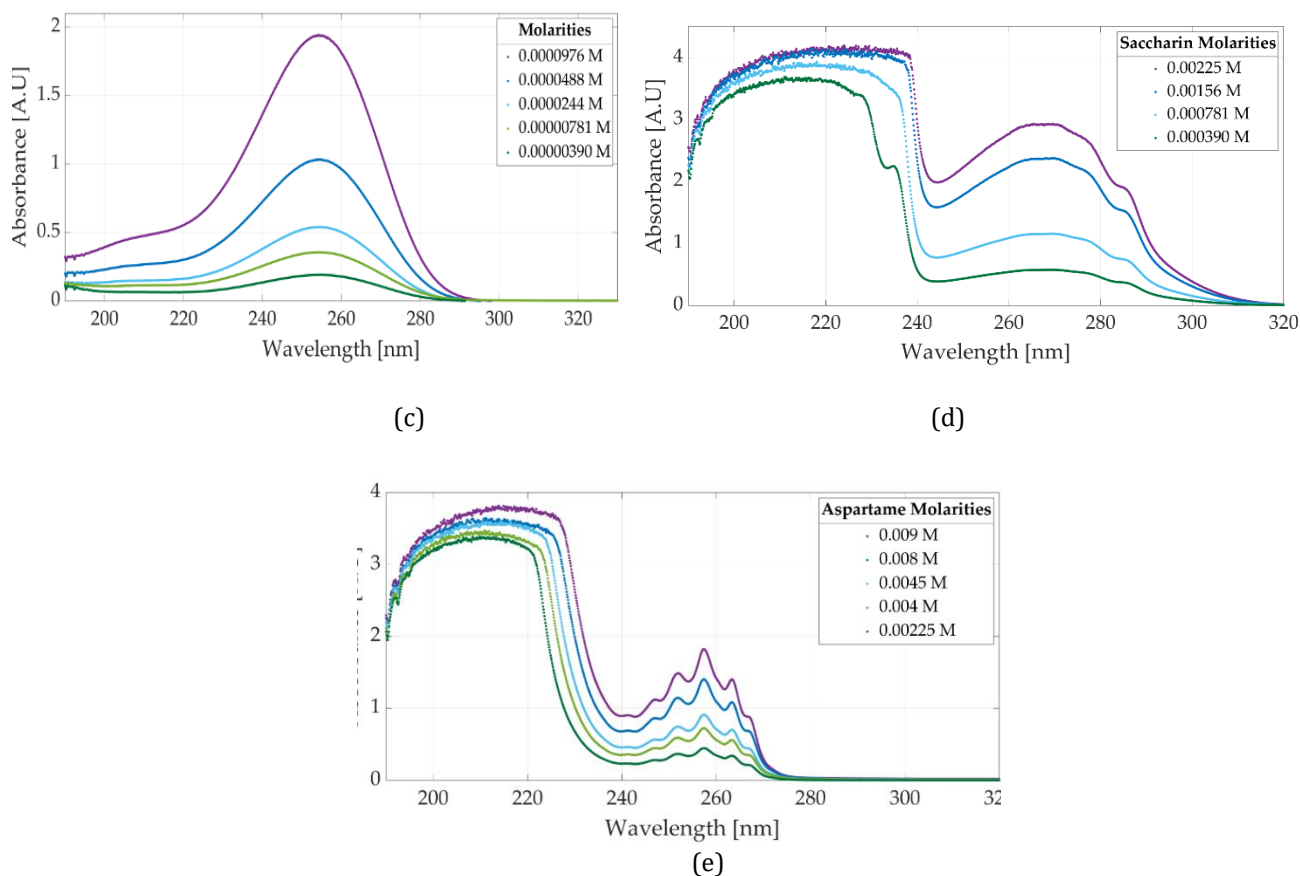
(c)

(d)

(e)
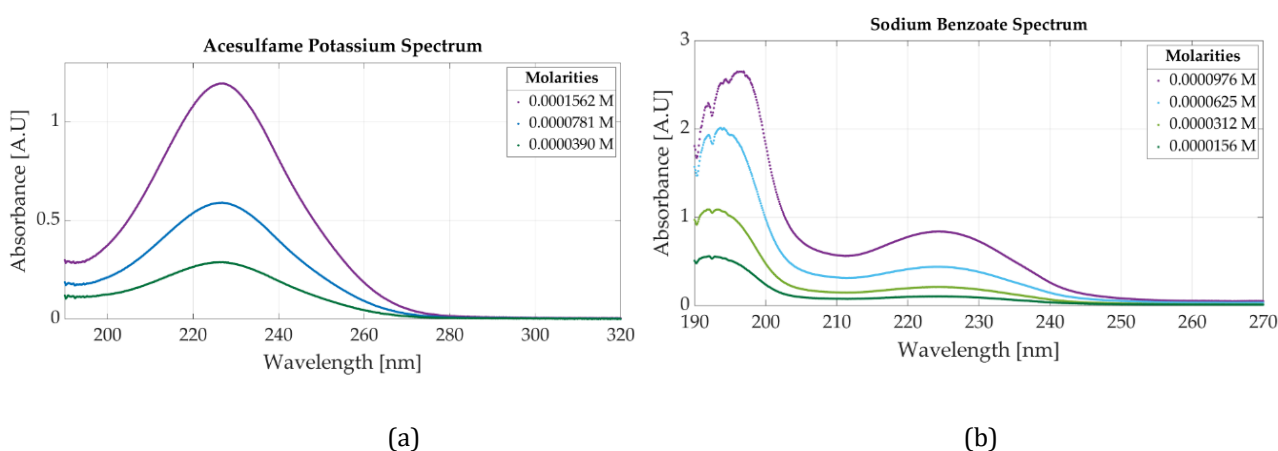
Figure 4.6 Spectra representation (a) for acesulfame potassium molarities; (b) for sodium benzoate molarities; (c) for potassium sorbate molarities; (d) for saccharin molarities; (e) for aspartame molarities [6]

Table 4.3 Spectral parameters for a sample of each additive

Parameters	Acesulfame potassium	Aspartame	Sodium benzoate	Potassium sorbate	Saccharin
Molar concentration [M]	0.000375	0.007	0.00025	0.00225	0.00225
Maximum absorbance [A.U]	3.494	1.629	1.673	3.392	2.929
λ_C [nm]	226.6	257.4	224	253.9	269.6
λ_L [nm]	207.7	242.8	211.2	232.1	244.3
λ_R [nm]	247	266.1	237	271.9	286.5
FWHM [nm]	39.3	23.3	25.8	39.8	42.2

Fifteen spectra were acquired for each of the five additives. For all seventy-five spectra the mentioned parameters were calculated. The data was used to train and test three machine learning techniques. The database was divided into two datasets: 80% was allocated into the training dataset, whereas the remaining 15 spectra were used to calculate the accuracy of the classification for testing.

The first technique used for the spectra classification was SVM. Each additive was given a numerical value, which appeared also in the confusion matrixes. The codification was the following: 1 for acesulfame potassium, 2 for aspartame, 3 for sodium benzoate, 4 for potassium sorbate and 5 for saccharin. Figure 4.8 (a) shows the classification results grouped within a confusion matrix. Horizontally are displayed the real classes of the solutions, whereas vertically are placed the predicted classes. The values within the diagonal indicate that all the samples were correctly identified by the techniques.
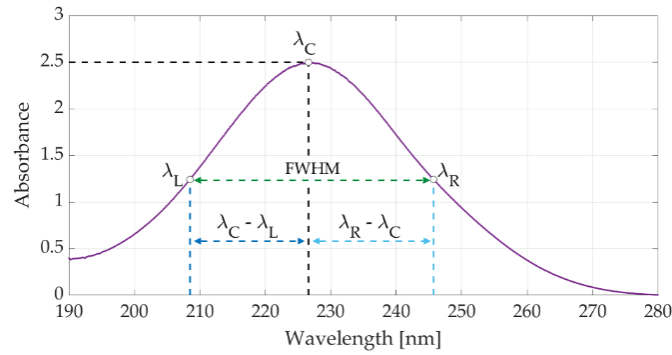
Figure 4.7 Spectral parameters

The second machine learning technique that was tested was tested was k-NN. The confusion matrix for this technique is illustrated in Figure 4.8 (b). The fifteen recordings were correctly identified with this technique too, the testing accuracy being 100%. Lastly, decision trees were used for the spectra classification. The confusion matrix within Figure 4.8 (c) shows that one sample from class 1 (acesulfame potassium) was incorrectly classified as a sample from class 3 (sodium benzoate). The analysis has shown that the most relevant parameters used in the classification of the data are λ_L, the relative error for λ_L, λ_C and Kurtosis.
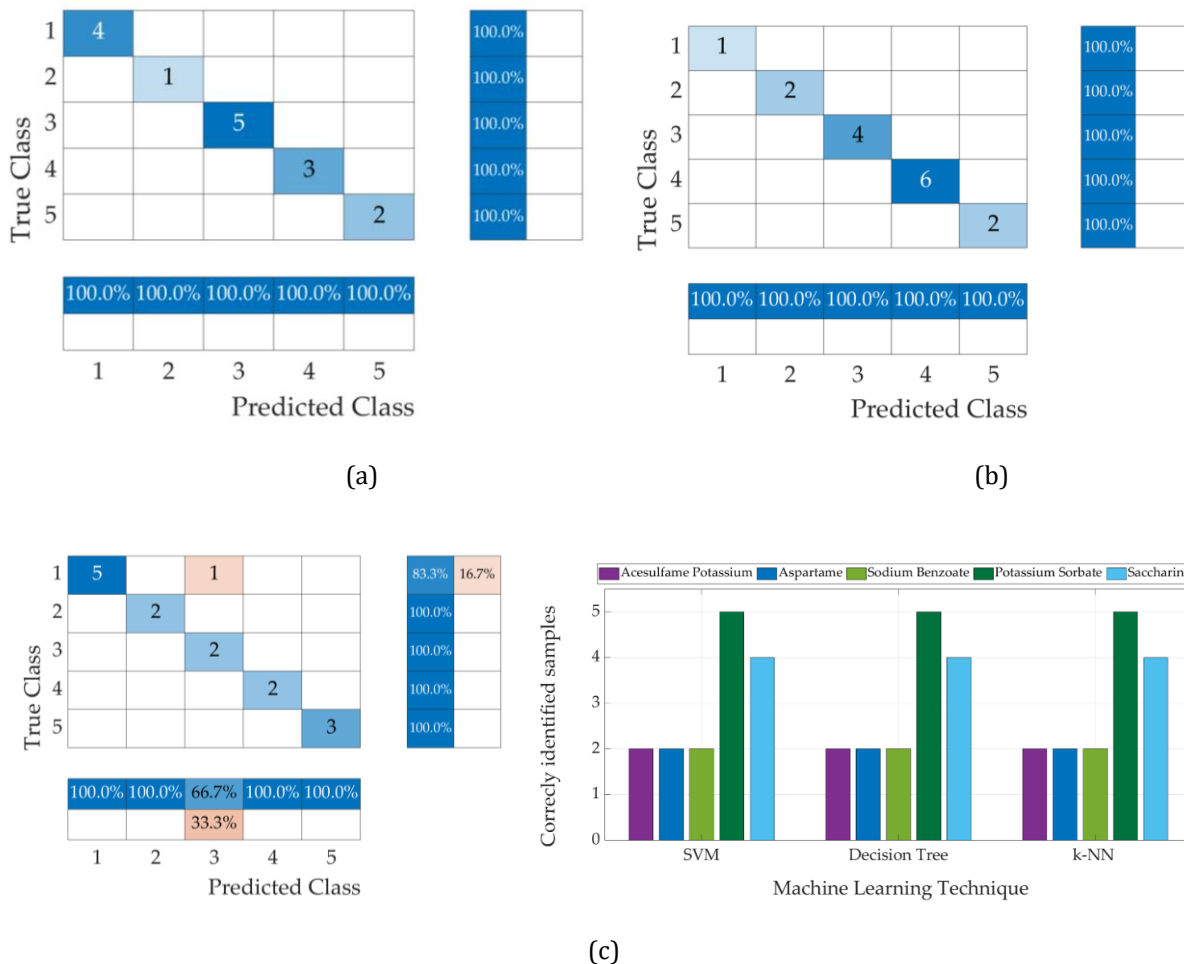

(a)

(b)

(c)

Figure 4.8 Confusion matrix (a) SVM algorithm; (b) kNN algorithm; (c) decision tree algorithm (d) classification results for validation

The mean testing classification accuracy was calculated by repeating the classification process ten times. The data was randomly divided into two datasets. The results showed that

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SVM and k-NN methods were capable of correctly identifying all the samples obtaining 100% accuracy, whereas with decision trees the mean accuracy was 99.33% ± 2.1 %.

A validation model was created for each technique. To determine the validation accuracy of the models an additional dataset containing fifteen recordings was used. The dataset contained the spectra of two acesulfame potassium, 2 aspartame, 2 sodium benzoate, 5 potassium sorbat and 4 saccharin samples. The results are presented in Figure 4.8 (d). The validation database contained few spectra; therefore, the algorithms were able to identify all the spectra correctly.

4.2.3   Deep learning techniques for food additive classification

The study of food additives was continued in chapter 6. It was desired to enlarge the spectra database for all five additives and also acquire and analyze the spectra of mixtures between two additives. There may be more additives within a sample; therefore, the goal was to assess if the spectra of mixed solutions (containing two additives) present the absorbance peaks of both additives, if the peaks overlapped and if the spectra of mixtures could be classified.

The molar concentration range of the simple solutions was extended, new molar concentrations were prepared and analyzed with the UV spectrometer. The final database for pure solutions was enlarged from 75 to 193 spectra. The study was continued with the preparation of mixed solutions. Simple solutions were prepared and 2 ml from each sample was mixed into a clean Berzelius glass. To obtain more spectra for the same mixture, but with different absorbance values, the solutions were diluted by adding 1.5 ml of distillate water with 1.5 ml of the sample. With the five additives ten mixed solutions were prepared and analyzed [6].

In Figure 4.6 (a) and (c) was shown that the absorbance peak of acesulfame potassium is located at 226 nm and at 254 nm for solutions of potassium sorbate. Figure 4.9 (a) depicts the spectra of three samples: the first spectrum corresponds to an acesulfame potassium solution of 0.00005 M; the second contains 0.0000156 M potassium sorbate, whereas the third sample is a mixture of acesulfame potassium and potassium sorbate. The molar concentration of the sample is 0.0000585 M acesulfame potassium and 0.0000219 M potassium sorbate. The mixture's spectrum shows the absorbance peak for potassium sorbate (at 254 nm) and the one for acesulfame potassium (at 226 nm) [6].
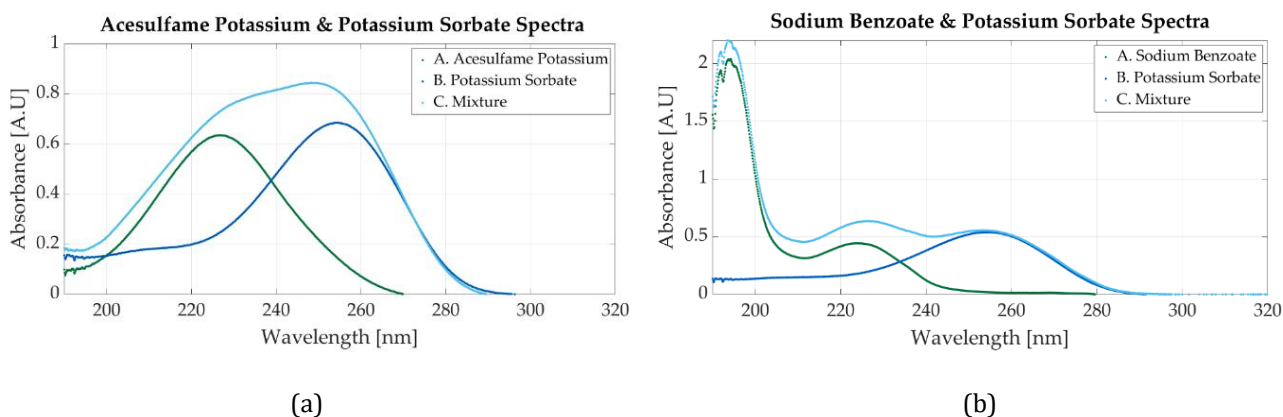

| (a) | (b) |

Figure 4.9 Additive spectra (a) A. Acesulfame potassium; B. Potassium sorbate; C. Mixture of acesulfame potassium and potassium sorbate; (b) A. Sodium benzoate; B. Potassium sorbate; C. Mixture of sodium benzoate and potassium sorbate [6]

Figure 4.9 (b) presents the spectrum of a mixture of sodium benzoate and potassium sorbate. The spectrum contains both absorbance peaks, which indicate the presence of both

additives within the sample. The peaks associated with sodium benzoate are located at 224 nm and 194 nm, whereas the one for potassium sorbate is visible at 254 nm [6]. Similar results were obtained for the other eight mixtures. The mixture database contained 211 spectra

The significant growth of the spectra database (404 spectra, 193 for pure solutions and 211 for mixtures of two additives) allowed the use of neural networks instead of the machine learning techniques for the sample classification. Two types of neural networks were used: artificial neural networks and convolutional neural networks. Figure 4.10 shows the architecture of the ANN with one hidden layer and with two hidden layers. The accuracy of the two architectures was compared.

The classification of the spectra was also done with CNN with 1, 2 and 3 convolutional layers (followed or not by pooling layers). The architecture of the CNN with three convolutional layers followed by pooling layers is illustrated in Figure 4.11. These networks were used for the classification of pure samples, mixture and for all the samples. Each algorithm was used ten times to calculate the mean testing/validation accuracy. For each classification, the data was randomly divided into datasets.

Table 4.4 presents the best accuracy results obtained by classifying the samples with the eight networks. The results show that CNN architectures can correctly identify more spectra and as a result have higher accuracy values (over 90%) compared to ANN [6]. An analysis of variance (ANOVA) was also performed for the accuracy datasets to determine if there is a statistical difference between performances of the networks. The results of the one-way ANOVA have also highlighted that CNN are more prone to obtain better classification results than ANN [6].
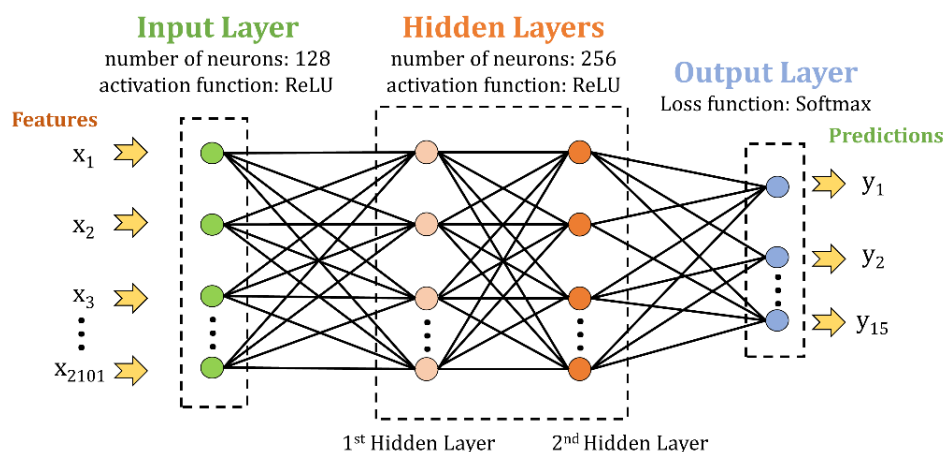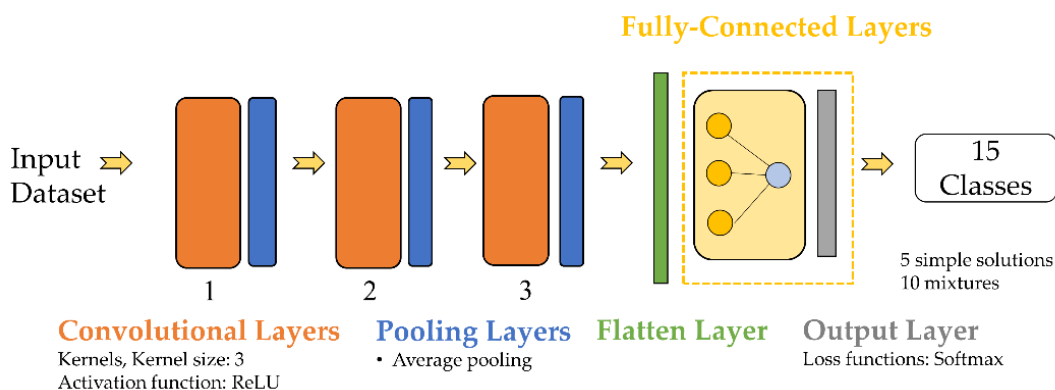

Figure 4.10 ANN architecture [6]

Figure 4.11 CNN architecture [6]

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Table 4.4 The accuracy of the models [6]

Dataset	Neural Network	Stage	Mean Accuracy [%]	Standard deviation [%]
Pure solutions	CNN2*	Testing	99.67	1.02
		Validation	99.67	1.02
Mixtures	CNN1*	Testing	91.94	4.02
		Validation	94.16	3.05
All samples	CNN3	Testing	92.38	1.48
		Validation	93.43	2.01

CNN2* - CNN with 2 convolutional layers and pooling layers
CNN1* - CNN with 1 convolutional layers and 1 pooling layer
CNN3* – CNN with 3 convolutional layers and 2 pooling layers

The results of this study showed that the classification of food additive can be done using UV spectroscopy and neural networks. A research paper utilized BP-ANN (back-propagation neural network algorithm) and PLS (partial least squares) for the analysis of the UV spectra of two food additives (sodium benzoate and potassium sorbate) [72]. These techniques were able to identify the two additives within simple solutions, within mixtures but also in real samples. This study has focused on the classification of the spectra of five additive, from two different classes (sweeteners and preservatives) and has also included the classification of ten different mixed solutions (two-additive solutions). This study has not assessed real samples, but the aim is to continue the research by acquiring the spectra of mixtures of three, four and even five additives. The addition of new food additives would also help in the analysis of real samples, because there may be samples that contain certain additives (which might not be included in this study) and without knowing the spectra they would be hard to identify.

4.2.4 Holographic sensors for acetic acid and ethyl alcohol detection

The analysis of food additives was continued in **chapter 7** by using holographic sensors. To record the holograms, it was desired to use a commercial photopolymer to reduce the preparation time. The disadvantage of using a commercial photopolymer is not knowing to which substances it may be sensitive or not. It was necessary to test the hologram with various analytes to determine which determine an optical response.

The holograms were recorded in a commercial photopolymer called Bayfol HX200 [73], [74]. Figure 4.12 and Figure 4.13 illustrate the setup used to record the holograms. The wavelength of the LASER is 532 nm. The photosensitive material was attached to a microscope slide to avoid its movement during the recording. On a single microscope slide, three holograms were recorded, as can be seen in Figure 4.14.
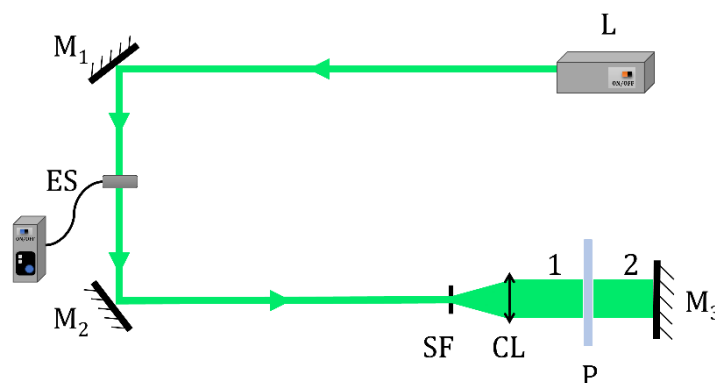

Figure 4.12 Recording setup (M_1, M_2, M_3 - mirrors, ES – electronic shutter, SF – spatial filter, CL – collimating lens, P – photopolymer, 1 – reference wave, 2 – object wave)[1] [75]

The recorded holograms were meant for the detection of food additives. The holographic sensors are sensitive to one or more additives if there is a displacement of the diffracted spectrum. Therefore, the spectrum needed to be monitored. Figure 4.15 (a) visually explains the wavelength displacement of the diffracted spectrum, whereas Figure 4.15 (a) shows the monitoring setup used for the holograms. The main components are a white light source, the cuvette with the sample, the holographic sensors (that is attached to one of the walls of the cuvette) and the spectrophotometer.
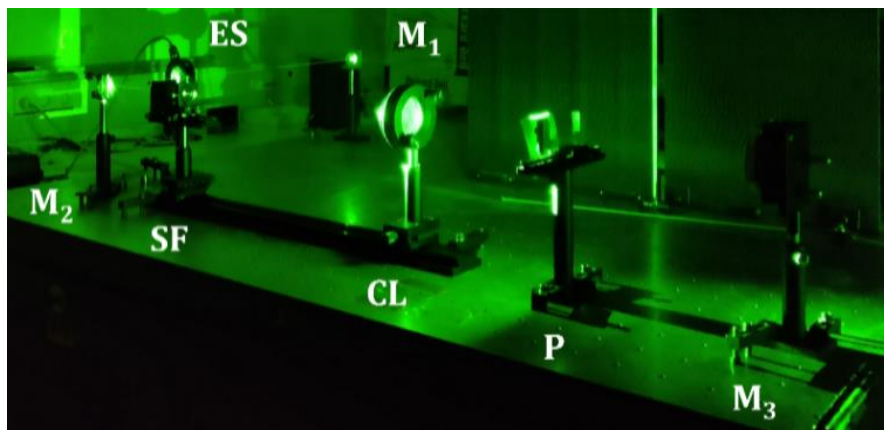

Figure 4.13 Real recoding setup (M_1, M_2, M_3 - mirrors, ES – electronic shutter, SF – spatial filter, CL – collimating lens, P – photopolymer) [75]
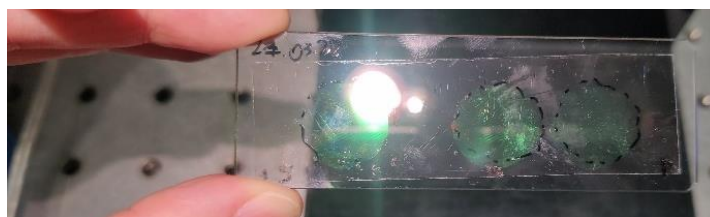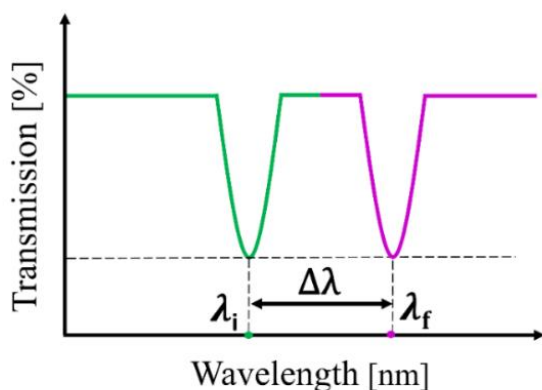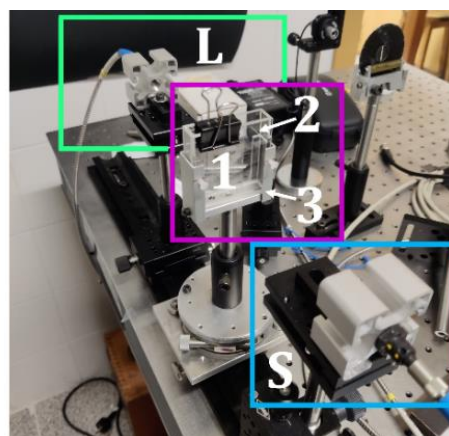

Figure 4.14 Microscope slide with three holograms

(a)   (b)

Figure 4.15 (a) Wavelength displacement of the diffracted spectrum (λ_i – initial wavelength; λ_f– final wavelength; $\Delta\lambda$ - wavelength displacement); (b) Wavelength displacement monitoring setup (L -tungsten halogen lamp; 1 – microscope slide with hologram;   2 – cuvette; 3 – cuvette support; S – spectrophotometer)[1] [75]

The holograms were tested with various food additives. In literature, holographic sensors were used for pH detection [37], [38]. Therefore, it was important to test if the

holographic sensors recorded on Bayfol could be used to determine the pH of a sample. The holograms were also tested with acid and bases (strong and weak) and with a type of alcohol.

The holograms were tested with sulfuric acid (strong acid), with citric acid, acetic acid and ascorbic acid), with sodium hydroxide (strong base), sodium bicarbonate (weak base) and ethanol (alcohol). Solutions with various concentrations were prepared and the wavelength displacement was monitored for 5 minutes. The results showed that there is no displacement of the diffracted spectrum for ascorbic acid, citric acid, sulfuric acid and for the two bases. Therefore, it was proven that these sensors cannot detect pH.

The results obtained for the ethanol and acetic acid solutions have shown that there is a dependence between the concentration of the samples and the wavelength displacement of the diffracted spectrum. Figure 4.16 shows the wavelength displacement for two acetic acid solutions. In the left part of the image was placed the wavelength displacement obtained for each solution. The sample with a molarity of 0.5 M had a wavelength displacement of 3.61 nm, whereas for the second (0.7 M) the displacement has grown to 4.7 nm.
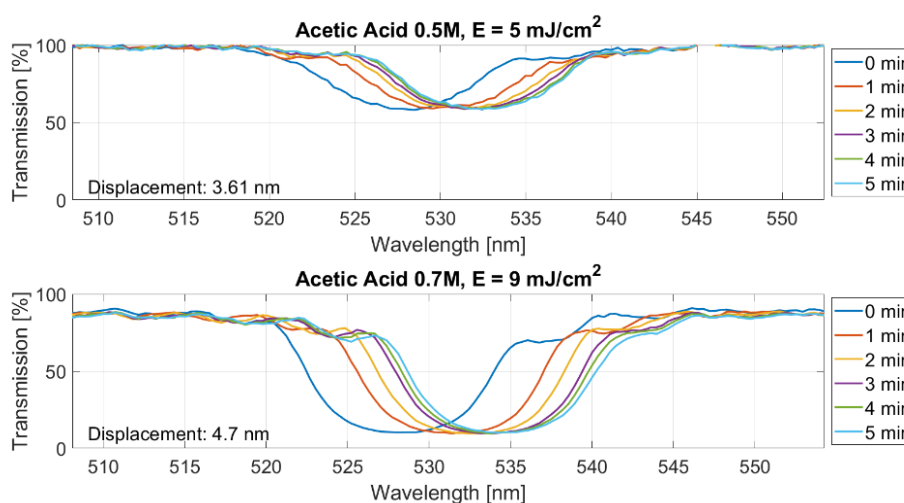

Figure 4.16 Experimental results for acetic acid solutions [1][75]

Figure 4.16 shows the wavelength displacement for the two samples of ethanol with concentrations of 50% (v/v) and 80% (v/v). Comparing the figures, it can be observed that the ethanol sample with lower concentration has a smaller wavelength displacement than the sample with 80% (v/v). More samples were prepared using ethanol and acetic acid. The ethanol concentration varied between 0-100% (v/v), whereas the ones for acetic acid varied between 0.004 M and 4 M.
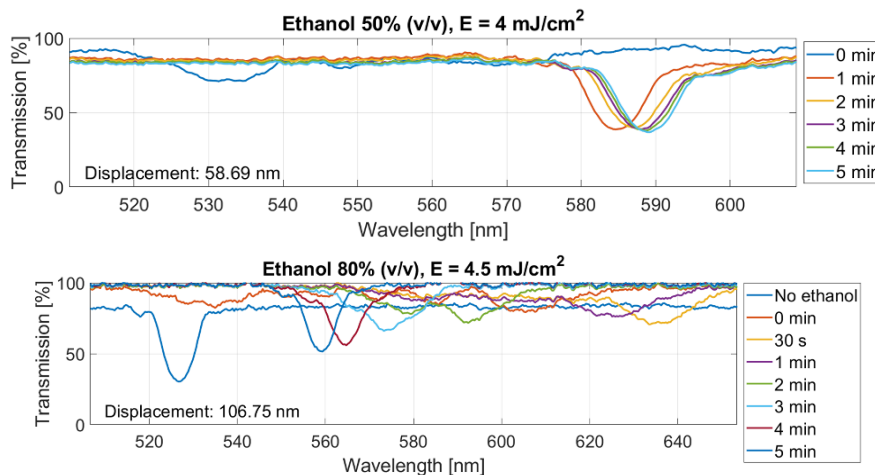

Figure 4.17 Experimental results for ethanol solutions 50% (v/v) and 80% (v/v)[1] [75]

The acetic acid measurements showed a growth in the wavelength displacement for molar concentration higher than 0.09 M after 5 minutes of analysis. For smaller concentrations of acetic acid, the response of the sensors fluctuated. The ethanol measurements showed a rise in the wavelength displacement for all concentration. The results obtained in Figure 4.16 show that the maximum displacement was obtained after 5 minutes for solutions of acetic acid. For ethyl alcohol samples, Figure 4.17, the maximum displacement is obtained after 1 minute for concentrations lower than 70% (v/v) and 30 seconds for higher concentrations.

Figure 4.18 (a) and (b) show the fitting curve obtained by representing the wavelength displacement and the molar concentration of the analyzed samples. The acetic acid results show that the two parameters depend according to a quadratic equation, $y = 0.3795 x^2 + 6.517x + 2.692$, whereas for the ethanol samples, with concentration higher than 70%, the equation is $y = 0.01014 x^2 + 0.5881x + 0.3587$.
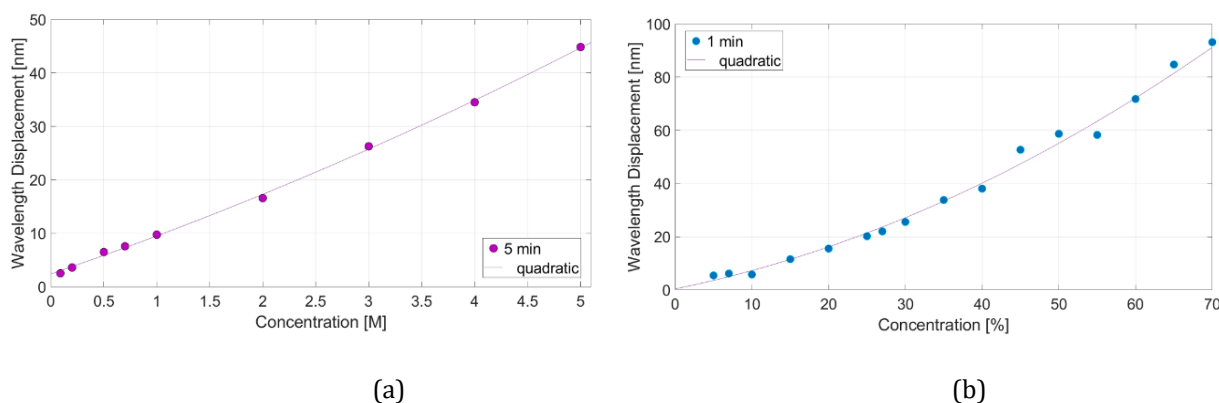

(a)   (b)

Figure 4.18 Fitting curve (a) for acetic acid; (b) for ethanol[1] [75]

In this study the concentration of acetic and ethyl alcohol samples was determined using holographic sensors recorded on a commercial photopolymer. The response time was 5 minutes for solutions of acetic acid and 1 minute for ethyl alcohol solution with concentrations lower than 70% (v/v). No holographic sensors for acetic acid detection were found in literature, but various methods (used for the analysis of acetic acid solutions) had response times varying from few seconds to minutes [76], [77]. Holographic sensor for ethanol detection responded to the presence of the analyte in 50 seconds for concentrations of 10% (v/v) [41], which is a very similar result compared to the one obtained in this study with the commercial photopolymer. Comparing the performance of these sensors with the ones obtained with other holographic sensors (which detect other substances and have response time up to 20 minutes [78]) it can be said that these sensors have a fast response time to the analytes.

5. Conclusions

This thesis has focus on the study of optical sensors and advanced methods that use light radiation for the analysis of drinks and food additives. The analysis methods based on light radiation for probe characterization are extremely used at the moment. This is confirmed by the large number of articles published in the last years using the following techniques: UV spectroscopy [21], [43], [79], infrared spectroscopy [59], [80], [81], fluorescence spectroscopy [82]–[84], interferometry [24], [85]–[88] and colorimetry [89].

In **chapter 1** a theoretical study was realized to better understand how optical sensors and analysis methods function. The study was concluded by identifying applications where optical sensors or optical analysis methods were used [16], [82], [90], [91].

In **chapter 2** machine learning and deep learning methods were discussed. Three machine learning algorithms were briefly discussed (k-NN, SVM, decision tree) followed by the presentation of the architecture of artificial and convolutional neural networks [52], [53]. These techniques were also used with optical analysis methods[58], [59].

Chapter 3 includes a short introduction on the thesis thematic, highlighting the main objectives. **Chapters 4-7** include the four-research direction that were studied during the PhD period: milk and juices analysis using low-cost optical devices; the classification of pure solutions of food additives using machine learning techniques; the classification of pure and mixed solutions of food additives using neural networks; acetic acid and ethanol detection using holographic sensor recorded on commercial photopolymer. Lastly, **chapter 8** presents the thesis conclusions and the personal contributions.

6. Personal contributions

- Testing experimental setups based on low optical devices.
- Analyzing milk and juice samples using low-cost experimental setups.
- Feature extraction for the UV spectra of five food additives (aspartame, acesulfame potassium, saccharin, sodium benzoate and potassium sorbate).
- Classification of the UV spectra (simple solutions of aspartame, acesulfame potassium, saccharin, sodium benzoate and potassium sorbate) using machine learning techniques.
- Spectra acquisition for 5 food additives: aspartame, acesulfame potassium, saccharine, sodium benzoate and potassium sorbate, obtaining a total of 193 UV spectra.
- Spectra acquisition for mixtures of two additives (211 UV spectra for mixtures)
- Study and testing of artificial neural network with one and two hidden layers for the classification of the UV spectra of food additives.
- Study and testing of convolutional neural network with one, two and three convolutional layers (preceded or not by pooling layers) for the classification of the UV spectra of food additives.
- The determination of the chemical species that inflict a wavelength displacement of the diffracted spectra in the holographic sensors recorded on a commercial photopolymer.
- The usage of a holographic sensor recorded on commercial photopolymer for the detection of acetic acid concentrations.
- The usage of a holographic sensor recorded on commercial photopolymer for the detection of ethyl alcohol concentrations.
- Testing holographic sensors recorded on commercial with various bases and acids to determine if the sensors are sensitive to pH (non-responsive to pH variations).
- Obtaining a dependency between the optical response of the holographic sensors recorded on a commercial photopolymer and the concentration of the samples.
- Obtaining a holographic sensor with fast response to analyte presence (5 minutes for acetic acid samples and 1 minute for ethanol concentration between 5% and 70% (v/v), respectively, 30 second for higher concentrations of ethanol.

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[1]The figures are based on the results obtained in: I-A. Potărniche, J. Marín-Sáez, M.-V. Collados, J. Atencia, "Holographic sensor based on Bayfol® commercial photopolymer for ethanol and acetic acid detection", which at the moment of submission of this resume (September 2023) was under review at the journal "Optics and Laser Technology".

7. Bibliography

[1]   K. W. Alt, A. Al-Ahmad, and J. P. Woelber, "Nutrition and Health in Human Evolution–Past to Present," *Nutrients*, vol. 14, no. 17, p. 3594, Aug. 2022, doi: 10.3390/nu14173594.

[2]   O. Gulsia, "Vanillin: One Drug, Many Cures," *Resonance*, vol. 25, no. 7, pp. 981–986, Jul. 2020, doi: 10.1007/s12045-020-1013-z.

[3]   S. A. A. Shaher, D. F. Mihailescu, and B. Amuzescu, "Aspartame Safety as a Food Sweetener and Related Health Hazards," *Nutrients*, vol. 15, no. 16, p. 3627, Aug. 2023, doi: 10.3390/nu15163627.

[4]   "FAO & WHO Codex Alimentarius, 'General Standard For Food Additives.'" https://www.fao.org/fao-who-codexalimentarius/sh-proxy/en/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXS%2B192-1995%252FCXS_192e.pdf (accessed Aug. 23, 2023).

[5]   "FAO & WHO Codex Alimentarius, 'Class Names And The International Numbering System For Food Additives.'" https://www.fao.org/fao-who-codexalimentarius/sh-proxy/en/?lnk=1&url=https%253A%252F%252Fworkspace.fao.org%252Fsites%252Fcodex%252FStandards%252FCXG%2B36-1989%252FCXG_036e.pdf (accessed Aug. 23, 2023).

[6]   I.-A. Potărniche, C. Saroşi, R. M. Terebeş, L. Szolga, and R. Gălătuş, "Classification of Food Additives Using UV Spectroscopy and One-Dimensional Convolutional Neural Network," *Sensors*, vol. 23, no. 17, p. 7517, Aug. 2023, doi: 10.3390/s23177517.

[7]   " Evaluations of the Joint FAO/WHO Expert Committee on Food Additives (JECFA)." https://apps.who.int/food-additives-contaminants-jecfa-database/ (accessed Aug. 23, 2023).

[8]   Y.-C. Chen *et al.*, "Aspartame Consumption, Mitochondrial Disorder-Induced Impaired Ovarian Function, and Infertility Risk," *Int J Mol Sci*, vol. 23, no. 21, p. 12740, Oct. 2022, doi: 10.3390/ijms232112740.

[9]   P. J. Landrigan and K. Straif, "Aspartame and cancer – new evidence for causation," *Environmental Health*, vol. 20, no. 1, p. 42, Dec. 2021, doi: 10.1186/s12940-021-00725-y.

[10]   C. Debras *et al.*, "Artificial sweeteners and cancer risk: Results from the NutriNet-Santé population-based cohort study," *PLoS Med*, vol. 19, no. 3, p. e1003950, Mar. 2022, doi: 10.1371/journal.pmed.1003950.

[11]   H. Rodrigues, C. Silva, and F. Martel, "The effects of aspartame on the HTR8/SVneo extravillous trophoblast cell line," *Reprod Biol*, vol. 22, no. 3, p. 100678, Sep. 2022, doi: 10.1016/j.repbio.2022.100678.

[12]   C.-H. Lin, H.-Y. Li, S.-H. Wang, Y.-H. Chen, Y.-C. Chen, and H.-T. Wu, "Consumption of Non-Nutritive Sweetener, Acesulfame Potassium Exacerbates Atherosclerosis through Dysregulation of Lipid Metabolism in ApoE−/− Mice," *Nutrients*, vol. 13, no. 11, p. 3984, Nov. 2021, doi: 10.3390/nu13113984.

[13]   A. Singh, K. Rourk, A. Bernier, and G. de Lartigue, "Non-Nutritive Sweetened Beverages Impair Therapeutic Benefits of Metformin in Prediabetic Diet-Induced Obese Mice," *Nutrients*, vol. 15, no. 11, p. 2472, May 2023, doi: 10.3390/nu15112472.

[14]   M. Urbano, M. D. Luque de Castro, P. M. Pérez, J. García-Olmo, and M. A. Gómez-Nieto, "Ultraviolet–visible spectroscopy and pattern recognition methods for differentiation and classification of wines," *Food Chem*, vol. 97, no. 1, pp. 166–175, Jul. 2006, doi: 10.1016/j.foodchem.2005.05.001.

[15]   S. Hao *et al.*, "Authentication of acacia honey using fluorescence spectroscopy," *Food Control*, vol. 130, p. 108327, Dec. 2021, doi: 10.1016/j.foodcont.2021.108327.

[16]   J. M. Pereira, L. M. Leme, M. R. F. G. Perdoncini, P. Valderrama, and P. H. Março, "Fast Discrimination of Milk Contaminated with Salmonella sp. Via Near-Infrared Spectroscopy," *Food Anal Methods*, vol. 11, no. 7, pp. 1878–1885, Jul. 2018, doi: 10.1007/s12161-017-1090-0.

[17] T. Desquins, F. Bousefsaf, A. Pruski, and C. Maaoui, "A Survey of Photoplethysmography and Imaging Photoplethysmography Quality Assessment Methods," *Applied Sciences*, vol. 12, no. 19, p. 9582, Sep. 2022, doi: 10.3390/app12199582.

[18] A. Breitinger, E. Clua, and L. A. F. Fernandes, "An Augmented Reality Periscope for Submarines with Extended Visual Classification," *Sensors*, vol. 21, no. 22, p. 7624, Nov. 2021, doi: 10.3390/s21227624.

[19] H. Zhang, H. Liu, W. Xu, and Z. Lu, "Large aperture diffractive optical telescope: A review," *Opt Laser Technol*, vol. 130, p. 106356, Oct. 2020, doi: 10.1016/j.optlastec.2020.106356.

[20] F. Massoumian, R. Juskaitis, M. A. A. Neil, and T. Wilson, "Quantitative polarized light microscopy," *J Microsc*, vol. 209, no. 1, pp. 13–22, Jan. 2003, doi: 10.1046/j.1365-2818.2003.01095.x.

[21] H. D. Xie, L. J. Bu, X. W. Peng, and Z. X. Li, "Ultraviolet Spectroscopy Method for Classifying Vinegars," *Adv Mat Res*, vol. 346, pp. 865–874, Sep. 2011, doi: 10.4028/www.scientific.net/AMR.346.865.

[22] J. Sádecká and M. Jakubíková, "Classification of Tokaj Wines by Ultraviolet–Visible Spectroscopy," *Food Anal Methods*, vol. 15, no. 1, pp. 56–66, Jan. 2022, doi: 10.1007/s12161-021-02097-y.

[23] A. Philippidis *et al.*, "Application of Ultraviolet-Visible Absorption Spectroscopy with Machine Learning Techniques for the Classification of Cretan Wines," *Foods*, vol. 10, no. 1, p. 9, Dec. 2020, doi: 10.3390/foods10010009.

[24] H. A. Razak, N. H. Sulaiman, H. Haroon, and A. S. Mohd Zain, "A fiber optic sensor based on Mach-Zehnder interferometer structure for food composition detection," *Microw Opt Technol Lett*, vol. 60, no. 4, pp. 920–925, Apr. 2018, doi: 10.1002/mop.31072.

[25] J. Park, "Optical Glucose Sensor Using Pressure Sensitive Paint," *Sensors*, vol. 21, no. 13, p. 4474, Jun. 2021, doi: 10.3390/s21134474.

[26] B. Kaur, S. Kumar, and B. K. Kaushik, "Novel Wearable Optical Sensors for Vital Health Monitoring Systems—A Review," *Biosensors (Basel)*, vol. 13, no. 2, p. 181, Jan. 2023, doi: 10.3390/bios13020181.

[27] W. Fan, K. A. Kam, H. Zhao, P. J. Culligan, and I. Kymissis, "An Optical Soil Sensor for NPK Nutrient Detection in Smart Cities," in *2022 18th International Conference on Intelligent Environments (IE)*, IEEE, Jun. 2022, pp. 1–4. doi: 10.1109/IE54923.2022.9826759.

[28] Y. Sun, G. I. N. Waterhouse, X. Qiao, J. Xiao, and Z. Xu, "Determination of chloramphenicol in food using nanomaterial-based electrochemical and optical sensors-A review," *Food Chem*, vol. 410, p. 135434, Jun. 2023, doi: 10.1016/j.foodchem.2023.135434.

[29] J. L. Santos and F. Farahi, *Handbook of optical sensors*. Crc Press, 2014.

[30] D.-W. Sun, *Infrared spectroscopy for food quality analysis and control*. Academic press, 2009.

[31] Y. Amakura, M. Okada, S. Tsuji, and Y. Tonogai, "High-performance liquid chromatographic determination with photodiode array detection of ellagic acid in fresh and processed fruits," *J Chromatogr A*, vol. 896, no. 1–2, pp. 87–93, Oct. 2000, doi: 10.1016/S0021-9673(00)00414-3.

[32] B. Guzowski and M. Łakomski, "Temperature Sensor Based on Periodically Tapered Optical Fibers," *Sensors*, vol. 21, no. 24, p. 8358, Dec. 2021, doi: 10.3390/s21248358.

[33] E. Maurice, G. Monnom, B. Dussardier, A. Saïssy, D. B. Ostrowsky, and G. W. Baxter, "Erbium-doped silica fibers for intrinsic fiber-optic temperature sensors," *Appl Opt*, vol. 34, no. 34, p. 8019, Dec. 1995, doi: 10.1364/AO.34.008019.

[34] J. Ascorbe, J. Corres, F. Arregui, and I. Matias, "Recent Developments in Fiber Optics Humidity Sensors," *Sensors*, vol. 17, no. 4, p. 893, Apr. 2017, doi: 10.3390/s17040893.

[35] P. J. Thomas and J. O. Hellevang, "A high response polyimide fiber optic sensor for distributed humidity measurements," *Sens Actuators B Chem*, vol. 270, pp. 417–423, Oct. 2018, doi: 10.1016/j.snb.2018.05.011.

[36] K. P. W. Dissanayake, W. Wu, H. Nguyen, T. Sun, and K. T. V. Grattan, "Graphene-Oxide-Coated Long-Period Grating-Based Fiber Optic Sensor for Relative Humidity and External

Refractive Index," *Journal of Lightwave Technology*, vol. 36, no. 4, pp. 1145–1151, Feb. 2018, doi: 10.1109/JLT.2017.2756097.

[37] A. J. Marshall, J. Blyth, C. A. B. Davidson, and C. R. Lowe, "pH-Sensitive Holographic Sensors," *Anal Chem*, vol. 75, no. 17, pp. 4423–4431, Sep. 2003, doi: 10.1021/ac020730k.

[38] K. Sharma, Heena, G. C. Mohanta, and R. Kumar, "Study of a pH-Sensitive Hologram for Biosensing Applications," in *HMAM2*, Basel Switzerland: MDPI, Mar. 2023, p. 22. doi: 10.3390/HMAM2-14272.

[39] S. Davies, Y. Hu, J. Blyth, N. Jiang, and A. K. Yetisen, "Reusable Dual-Photopolymerized Holographic Glucose Sensors," *Adv Funct Mater*, vol. 33, no. 18, May 2023, doi: 10.1002/adfm.202214197.

[40] G. Khalili Moghaddam, H. Margerison, J. Suzuki, J. Blyth, and C. R. Lowe, "A transparent glucose-sensitive double polymerised holographic sensor," *Sens Actuators B Chem*, vol. 267, pp. 1–4, Aug. 2018, doi: 10.1016/j.snb.2018.04.009.

[41] A. K. Yetisen, I. Naydenova, F. da Cruz Vasconcellos, J. Blyth, and C. R. Lowe, "Holographic Sensors: Three-Dimensional Analyte-Sensitive Nanostructures and Their Applications," *Chem Rev*, vol. 114, no. 20, pp. 10654–10696, Oct. 2014, doi: 10.1021/cr500116a.

[42] S. Ahuja and M. Dong, *Handbook of pharmaceutical analysis by HPLC*. Elsevier, 2005.

[43] J. Molina, A. Flores, A. Manzo Robledo, and J. M. Gutierrez, "Green and Black Tea Characterization By Electrochemical and UV-Vis Spectroscopy Techniques for Bioelectronic Applications," *ECS Trans*, vol. 110, no. 1, pp. 1–5, Feb. 2023, doi: 10.1149/11001.0001ecst.

[44] L. H. Nurani *et al.*, "Chemometrics-Assisted UV-Vis Spectrophotometry for Quality Control of Pharmaceuticals: A Review," *Indonesian Journal of Chemistry*, vol. 23, no. 2, p. 542, Mar. 2023, doi: 10.22146/ijc.74329.

[45] L.-M. Yuan *et al.*, "Consensual Regression of Soluble Solids Content in Peach by Near Infrared Spectrocopy," *Foods*, vol. 11, no. 8, p. 1095, Apr. 2022, doi: 10.3390/foods11081095.

[46] D. Shon, S. Park, S. Yoon, and Y. Ko, "Identification of Biochemical Differences in White and Brown Adipocytes Using FTIR Spectroscopy," *Applied Sciences*, vol. 12, no. 6, p. 3071, Mar. 2022, doi: 10.3390/app12063071.

[47] W. Miloua, M. Ortuño, V. Navarro-Fuster, A. Beléndez, and I. Pascual, "Adulterant Detection in Peppermint Oil by Means of Holographic Photopolymers Based on Composite Materials with Liquid Crystal," *Polymers (Basel)*, vol. 14, no. 5, p. 1061, Mar. 2022, doi: 10.3390/polym14051061.

[48] A. M. Mustafa *et al.*, "A new HPLC-MS/MS method for the simultaneous determination of 36 polyphenols in blueberry, strawberry and their commercial products and determination of antioxidant activity," *Food Chem*, vol. 367, p. 130743, Jan. 2022, doi: 10.1016/j.foodchem.2021.130743.

[49] M. Grembecka, P. Baran, A. Błażewicz, Z. Fijałek, and P. Szefer, "Simultaneous determination of aspartame, acesulfame-K, saccharin, citric acid and sodium benzoate in various food products using HPLC–CAD–UV/DAD," *European Food Research and Technology*, vol. 238, no. 3, pp. 357–365, Mar. 2014, doi: 10.1007/s00217-013-2111-x.

[50] S. Taha, A. Al-Kharosi, W. Al-Sabari, F. AL-shidhani, and S. Alfarsi, "A Simple Method for Simultaneous Determination of Commonly used Artificial Food Colors and Preservatives in Soda, Jam, and Yogurt by HPLC-PDA," *Brazilian Journal of Analytical Chemistry*, vol. 7, no. 26, 2020, doi: 10.30744/brjac.2179-3425.AR-23-2019.

[51] H. Tohma, İ. Gülçin, E. Bursal, A. C. Gören, S. H. Alwasel, and E. Köksal, "Antioxidant activity and phenolic compounds of ginger (Zingiber officinale Rosc.) determined by HPLC-MS/MS," *Journal of Food Measurement and Characterization*, vol. 11, no. 2, pp. 556–566, Jun. 2017, doi: 10.1007/s11694-016-9423-z.

[52] A. Zafar *et al.*, "A Comparison of Pooling Methods for Convolutional Neural Networks," *Applied Sciences*, vol. 12, no. 17, p. 8643, Aug. 2022, doi: 10.3390/app12178643.

[53] S. Cong and Y. Zhou, "A review of convolutional neural network architectures and their optimizations," *Artif Intell Rev*, vol. 56, no. 3, pp. 1905–1969, Mar. 2023, doi: 10.1007/s10462-022-10213-5.

[54] S.-Y. Hwang and J.-J. Kim, "A Universal Activation Function for Deep Learning," *Computers, Materials & Continua*, vol. 75, no. 2, pp. 3553–3569, 2023, doi: 10.32604/cmc.2023.037028.

[55] Y. Yu, K. Adu, N. Tashi, P. Anokye, X. Wang, and M. A. Ayidzoe, "RMAF: Relu-Memristor-Like Activation Function for Deep Learning," *IEEE Access*, vol. 8, pp. 72727–72741, 2020, doi: 10.1109/ACCESS.2020.2987829.

[56] H. Zhang, S. Wang, D. Li, Y. Zhang, J. Hu, and L. Wang, "Edible Gelatin Diagnosis Using Laser-Induced Breakdown Spectroscopy and Partial Least Square Assisted Support Vector Machine," *Sensors*, vol. 19, no. 19, p. 4225, Sep. 2019, doi: 10.3390/s19194225.

[57] K. Przybył, K. Koszela, F. Adamski, K. Samborska, K. Walkowiak, and M. Polarczyk, "Deep and Machine Learning Using SEM, FTIR, and Texture Analysis to Detect Polysaccharide in Raspberry Powders," *Sensors*, vol. 21, no. 17, p. 5823, Aug. 2021, doi: 10.3390/s21175823.

[58] P. Sun, J. Wang, and Z. Dong, "CNN–LSTM Neural Network for Identification of Pre-Cooked Pasta Products in Different Physical States Using Infrared Spectroscopy," *Sensors*, vol. 23, no. 10, p. 4815, May 2023, doi: 10.3390/s23104815.

[59] Y. Hao, X. Li, C. Zhang, and Z. Lei, "Online Inspection of Browning in Yali Pears Using Visible-Near Infrared Spectroscopy and Interpretable Spectrogram-Based CNN Modeling," *Biosensors (Basel)*, vol. 13, no. 2, p. 203, Jan. 2023, doi: 10.3390/bios13020203.

[60] T. Chen, L. Ma, Z. Tang, and L. X. Yu, "Identification of coumarin-based food additives using terahertz spectroscopy combined with manifold learning and improved support vector machine," *J Food Sci*, vol. 87, no. 3, pp. 1108–1118, Mar. 2022, doi: 10.1111/1750-3841.16064.

[61] L. A. Szolga and T. R. Cilean, "Nitrates and Nitrites Detection System in the Drinking Water Using UV Absorption," in *2020 International Conference on e-Health and Bioengineering (EHB)*, IEEE, Oct. 2020, pp. 1–4. doi: 10.1109/EHB50910.2020.9280233.

[62] L. A. Szolga and F. Mudure, "Blood (RBC/HGB) Analysis Using Colorimetry," in *2020 International Conference on e-Health and Bioengineering (EHB)*, IEEE, Oct. 2020, pp. 1–4. doi: 10.1109/EHB50910.2020.9280195.

[63] L. A. Szolga and T. R. Cilean, "Water Sterilization Using Power UV LEDs," in *2021 8th International Conference on Electrical and Electronics Engineering (ICEEE)*, IEEE, Apr. 2021, pp. 49–53. doi: 10.1109/ICEEE52452.2021.9415965.

[64] L. A. Szolga, "Humidity and Isopropyl Alcohol Detection Sensor Based on Plastic Optical Fiber," in *2021 25th International Conference Electronics*, IEEE, Jun. 2021, pp. 1–6. doi: 10.1109/IEEECONF52705.2021.9467472.

[65] I.-A. Potarniche and R.-V. Galatus, "Spectrometric Milk Analyzer," in *2021 IEEE 27th International Symposium for Design and Technology in Electronic Packaging (SIITME)*, IEEE, Oct. 2021, pp. 214–217. doi: 10.1109/SIITME53254.2021.9663432.

[66] "AMS, AS7262 6-Channel Visible Spectral_ID Device with Electronic Shutter and Smart Interface, AS7262 Datasheet, [Revised March 2017]." Accessed: Jul. 05, 2023. [Online]. Available: https://www.mouser.com/datasheet/2/588/AS7262_DS000486_2-00-1082195.pdf

[67] "MulticompPRO, LED Pure White, 5mm, LED Datasheet, [Revised Aug. 2019]." Accessed: Jul. 05, 2023. [Online]. Available: https://www.farnell.com/datasheets/2861562.pdf

[68] R. Irkin, S. Dogan, N. Degirmencioglu, M. Diken, and M. Guldas, "Phenolic content, antioxidant activities and stimulatory roles of citrus fruits on some lactic acid bacteria," *Arch Biol Sci*, vol. 67, no. 4, pp. 1313–1321, 2015, doi: 10.2298/ABS140909108I.

[69] K. Shaikh, A. Waqas, U. A. Korai Baloch, B. Muneer, and A. Memon, "Cost-Effective Portable Photonic Sensor for Liquid Adulteration Detection," in *2022 IEEE 19th International Conference on Smart Communities: Improving Quality of Life Using ICT, IoT and AI (HONET)*, IEEE, Dec. 2022, pp. 063–068. doi: 10.1109/HONET56683.2022.10019183.

[70] I. Potarniche, R. Galatus, and L. Szolga, "Fresh juice pH evaluation using colorimetry," in *Advanced Topics in Optoelectronics, Microelectronics, and Nanotechnologies XI*, M. Vladescu, I. Cristea, and R. D. Tamas, Eds., SPIE, Mar. 2023, p. 93. doi: 10.1117/12.2643291.

[71] "JASCO Inc.,Specifications - JASCO Spectrophotometer." Accessed: Jul. 05, 2023. [Online]. Available: https://jascoinc.com/products/spectroscopy/uv-visible-nir-spectrophotometers/specifications/

[72] M. Wang *et al.*, "Simultaneous ultraviolet spectrophotometric determination of sodium benzoate and potassium sorbate by BP-neural network algorithm and partial least squares," *Optik (Stuttg)*, vol. 201, p. 163529, Jan. 2020, doi: 10.1016/j.ijleo.2019.163529.

[73] "Covestro, Bayfol® HX200, Bayfol® HX200 Datasheet, [Revised March. 2018]." Accessed: Jul. 05, 2023. [Online]. Available: https://www.google.com/url?sa=t&rct=j&q=&esrc=s&source=web&cd=&cad=rja&uact=8&ved=2ahUKEwjq3bel_ff_AhW1hf0HHZYQBJMQFnoECBwQAQ&url=https%3A%2F%2Fsolutions.covestro.com%2F-%2Fmedia%2Fcovestro%2Fsolution-center%2Fproducts%2Fdatasheets%2Fimported%2Fbayfol%2Fbayfol-hx200_en_86194384-20033146-20033738.pdf&usg=AOvVaw2YvCEj0_PlfKFODuDw9jMb&opi=89978449

[74] F.-K. Bruder, T. Fäcke, and T. Rölle, "The Chemistry and Physics of Bayfol® HX Film Holographic Photopolymer," *Polymers (Basel)*, vol. 9, no. 12, p. 472, Sep. 2017, doi: 10.3390/polym9100472.

[75] Ioana-Adriana Potărniche, Julia Marín-Sáez, María-Victoria Collados, and Jesús Atencia, "Holographic sensor based on Bayfol HX200 commercial photopolymer for ethanol and acetic acid detection, Under Review, September 2023," *Opt Laser Technol*.

[76] L. Pont, J. Barbosa, and F. Benavente, "A rapid and simple method for the determination of organic acids in proteolytic enzymes by capillary electrophoresis with indirect ultraviolet detection," *Microchemical Journal*, vol. 158, p. 105204, Nov. 2020, doi: 10.1016/j.microc.2020.105204.

[77] M. Ji, Y. Zhong, M. Li, R. Tan, Y. Hu, and G. Li, "Determination of acetic acid in enzymes based on the cataluminescence activity of graphene oxide–supported carbon nanotubes coated with NiMn layered double hydroxides," *Microchimica Acta*, vol. 190, no. 6, p. 231, Jun. 2023, doi: 10.1007/s00604-023-05808-w.

[78] S. Davies *et al.*, "Holographic Sensors in Biotechnology," *Adv Funct Mater*, vol. 31, no. 47, Nov. 2021, doi: 10.1002/adfm.202105645.

[79] F. Hajirasouliha, H. Omidi, N. Jafary Omid, and F. Abedin Dorkoosh, "UV spectroscopy: A novel method for determination of degree of substitution of phthaloyl group as amine protector in chitosan," *Zeitschrift für Physikalische Chemie*, vol. 237, no. 6, pp. 663–673, Jun. 2023, doi: 10.1515/zpch-2021-3017.

[80] R. Joshi *et al.*, "Deep Learning-Based Quantitative Assessment of Melamine and Cyanuric Acid in Pet Food Using Fourier Transform Infrared Spectroscopy," *Sensors*, vol. 23, no. 11, p. 5020, May 2023, doi: 10.3390/s23115020.

[81] J. Liu, J. Zhang, Z. Tan, Q. Hou, and R. Liu, "Detecting the content of the bright blue pigment in cream based on deep learning and near-infrared spectroscopy," *Spectrochim Acta A Mol Biomol Spectrosc*, vol. 270, p. 120757, Apr. 2022, doi: 10.1016/j.saa.2021.120757.

[82] H. Bian *et al.*, "Application of fluorescence spectroscopy in classifying apple juice according to the variety," *Optik (Stuttg)*, vol. 231, p. 166361, Apr. 2021, doi: 10.1016/j.ijleo.2021.166361.

[83] F. Venturini, S. Fluri, and M. Baumgartner, "Dataset of Fluorescence EEM and UV Spectroscopy Data of Olive Oils during Ageing," *Data (Basel)*, vol. 8, no. 5, p. 81, Apr. 2023, doi: 10.3390/data8050081.

[84] P. Viñas, C. López-Erroz, J. J. Marín-Hernández, and M. Hernández-Córdoba, "Determination of phenols in wines by liquid chromatography with photodiode array and fluorescence detection," *J Chromatogr A*, vol. 871, no. 1–2, pp. 85–93, Feb. 2000, doi: 10.1016/S0021-9673(99)01087-0.

[85] X. Sun, H. Du, X. Dong, Y. Hu, and J. Duan, "Simultaneous Curvature and Temperature Sensing Based on a Novel Mach-Zehnder Interferometer," *Photonic Sensors*, vol. 10, no. 2, pp. 171–180, Jun. 2020, doi: 10.1007/s13320-019-0551-z.

[86] R. S. El Shamy, X. Li, M. M. ElRayany, A. Sultan, and M. A. Swillam, "On-Chip Loop-Terminated Mach-Zehnder Interferometer Gas Sensor," in *2022 Photonics North (PN)*, IEEE, May 2022, pp. 1–1. doi: 10.1109/PN56061.2022.9908408.

[87] Y. Zhao *et al.*, "Ultrasensitive temperature sensor with Vernier-effect improved fiber Michelson interferometer," *Opt Express*, vol. 29, no. 2, p. 1090, Jan. 2021, doi: 10.1364/OE.415857.

[88] P. Wang, K. Ni, B. Wang, Q. Ma, and W. Tian, "Methylcellulose coated humidity sensor based on Michelson interferometer with thin-core fiber," *Sens Actuators A Phys*, vol. 288, pp. 75–78, Apr. 2019, doi: 10.1016/j.sna.2019.01.031.

[89] H. Zhang, H. Yang, P. Liu, X. Qin, and G. Liu, "Colorimetric quantification of sodium benzoate in food by using d-amino acid oxidase and 2D metal organic framework nanosheets mediated cascade enzyme reactions," *Talanta*, vol. 237, p. 122906, Jan. 2022, doi: 10.1016/j.talanta.2021.122906.

[90] N. N. Bachok, N. Burham, A. R. Othman, N. H. Abd. Karim, A. A. bin Abu Bakar, and N. Arsad, "Ascorbic Acid Detection Using Spectrometer and Deuterium and Halogen Light Source," in *2022 IEEE International Conference on Semiconductor Electronics (ICSE)*, IEEE, Aug. 2022, pp. 81–84. doi: 10.1109/ICSE56004.2022.9863177.

[91] Faragó, Gălătuș, Hintea, Boșca, Feurdean, and Ilea, "An Intra-Oral Optical Sensor for the Real-Time Identification and Assessment of Wine Intake," *Sensors*, vol. 19, no. 21, p. 4719, Oct. 2019, doi: 10.3390/s19214719.

8. List of publications

The author has worked on 7 manuscripts during the PhD studies that began in October 2019.

Journal papers indexed ISI (WOS):

1. **Potărniche, I.-A.;** Saroşi, C.; Terebeş, R.M.; Szolga, L.; Gălătuş, R. Classification of Food Additives Using UV Spectroscopy and One-Dimensional Convolutional Neural Network. *Sensors* 2023, *23*, 7517. https://doi.org/10.3390/s23177517

Under review:

2. **Ioana-Adriana Potărniche**, Julia Marín-Sáez, María-Victoria Collados, and Jesús Atencia, "Holographic sensor based on Bayfol HX200 commercial photopolymer for ethanol and acetic acid detection, Optics and Laser Technology.

Conference papers indexed ISI (WOS):

3. **Potărniche, I. A**., & Gălătuş, R. V. (2021, October). Spectrometric Milk Analyzer. In 2021 IEEE 27th International Symposium for Design and Technology in Electronic Packaging (SIITME) (pp. 214-217). IEEE. **WOS**:000786441900051; **DOI**: 10.1109/SIITME53254.2021.9663432.

4. Szolga, L. A., Heredea, P. C., & **Potarniche, I. A**. (2021, October). Low-Cost Peristaltic Pump for Laboratory Applications. In 2021 IEEE 27th International Symposium for Design and Technology in Electronic Packaging (SIITME) (pp. 322-325). IEEE. **WOS**:000786441900076; **DOI**: 10.1109/SIITME53254.2021.9663609.

5. Buzura, L., Budileanu, M. L., **Potarniche, A**., & Galatus, R. (2021, October). Python based portable system for fast characterisation of foods based on spectral analysis. In 2021 IEEE 27th International Symposium for Design and Technology in Electronic Packaging (SIITME) (pp. 275-280). IEEE. WOS:000786441900066; DOI: 10.1109/SIITME53254.2021.9663677.

Conference papers indexed Scopus (SPIE):

6. **Potărniche, I. A**., Gălătuş, R., & Szolga, L. (2023, March). Fresh juice pH evaluation using colorimetry. In Advanced Topics in Optoelectronics, Microelectronics, and Nanotechnologies XI (Vol. 12493, pp. 227-231). SPIE. **DOI**: https://doi.org/10.1117/12.2643291

Novice Insights in Electronics and Telecommunications:

7. **Ioana-Adriana Potărniche**, Codruța Saroşi, Romulus Terebeş, Ramona Gălătuş, Lorant Szolga. "Citric and ascorbic acid classification system", Novice Insights in Electronics and Telecommunications, Student Symposium on Electronics and Telecommunications UTCN, 2023, pp. 74-75, ISSN: 1842-6085.

   The paper was awarded second prize at the Master/PhD section.

Other published paper:

8. Groza, R., **Potarniche, I. A.**, Kirei, B. S., & Topa, M. D. (2018). Digitally controlled oscillator for all-digital frequency locked loops. *Romanian Journal of Information Science and Technology*, *21*(1), 3-17.

9. V. -I. -M. Chereja, **A. -I. Potarniche**, S. -A. Ranga, B. S. Kirei And M. D. Topa, "Power Dissipation Estimation of CMOS Digital Circuits at the Gate Level in VHDL," 2018 International Symposium on Electronics and Telecommunications (ISETC), Timisoara, Romania, 2018, pp. 1-4, doi: 10.1109/ISETC.2018.8583957.

Novice Insights in Electronics and Telecommunications:

10. **Ioana-Adriana Potărniche**, Robert Groza. "Environmental monitoring and control system", Novice Insights in Electronics and Telecommunications, Student Symposium on Electronics and Telecommunications UTCN, 2019, pp. 74-75, ISSN: 1842-6085, https://etti.utcluj.ro/files/Acasa/Site/SSET/Brosura%20SSET-2019.pdf