

Bio-impedance and circuit parameters: An analysis for tracking fruit ripening

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ABSTRACT

The evaluation of fruit quality from the field to the table, through its storage, handling and transport has become of paramount importance to meet production and consumers demands. For this purpose, fast, reliable and low-cost non-destructive techniques are highly desirable, to avoid food waste and allow a real-time decision making. Among non-destructive techniques, Electrical Impedance Spectroscopy (EIS) has shown great potential due to the possibility to correlate the physio-chemical evolution of the fruit to changes of electrical parameters. In this paper, the effect of ageing on apples and bananas during 13 d at room temperature was studied using a microcontroller-based EIS system, in a frequency range from 100 Hz to 85 kHz.

The bio-impedance changes were evaluated over time and the influence of the applied frequencies on its variation was investigated. Data were fitted with a proposed equivalent circuit, modelling both the interaction between the fruit and the sensor and the flow of current in the samples tissues. To validate the results, the circuit parameter changes were physiologically explained and the fitting compared with models found in literature. The results highlighted the potential of this non-destructive technique for monitoring the ripening and senescence of fruit, obtaining a good correlation of the impedance evolution with the low frequency points. The model fitting resulted in a Root Mean Squared Error (RMSE), for apples (376.5 Ω - 2.66%) and bananas (110.8 Ω - 2.82%), was comparable or better than best literature models. Finally, changes of circuit component values over time was explained for the electrode-fruit interaction and for the current flow in the plant tissues, giving a better insight of fruit ripening and senescence.

1. Introduction

There is a growing need for monitoring of fruit quality throughout the production chain. However, the conventional methods used for fruit quality assessment are commonly time-consuming, destructive and since for statistical significance they require many samples taken, they generate a substantial waste of good fruit (Vanoli and Buccheri, 2012). The non-destructive evaluation of fruit ripeness and quality, compared to classical methods, gives the possibility of obtaining high throughput measurements, which can be carried out both on-plant and post-harvest, that are rapid, accurate, environmentally friendly and that allow real-time decision-making (Li et al., 2018).

Electrical Impedance Spectroscopy (EIS) has gained popularity as a reliable alternative to the classical non-destructive techniques (Muñoz-Huerta et al., 2014). EIS quantitatively characterizes the interaction between the dielectric dipole moment of a sample under test and an

externally applied electric field. In particular, the impedance value output of the EIS measurement is dependent on the frequency of the applied signal. The choice of the input frequency range is typically dependent on the structure and chemical composition of the studied sample, as the correlation of quality parameters with the measured impedance appears at specific frequencies for different types of materials (El Khaled et al., 2017). EIS techniques are applied for multiple applications, such as food products screening (Grossi et al., 2011), solid materials properties characterization (Iqbal and Rafiuddin, 2016) and human body analysis (Clemente et al., 2014).

The first studies on the use of EIS to characterize the physiological state of fruit were based on destructive methods. The most relevant studies were performed on apples (Jackson and Harker, 2000; Li et al., 2005), avocados (Montoya et al., 1994), persimmons (Harker and Forbes, 1997), nectarines (Harker and Mandonald, 1994), kiwifruit (Bauchot et al., 2000), pears (Montoya et al., 1994) and tomatoes

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(Benavente et al., 1998). More recently, non-destructive methods have been adopted for the estimation of the quality of large fruit numbers; for example, apples (Yovcheva et al., 2013), bananas (Chowdhury et al., 2015), lettuce (Muñoz-Huerta et al., 2014), mangoes (Rehman et al., 2011) and strawberries (González-Araiza et al., 2017). In apples and bananas, impedance measurements have been correlated with the ripening process (Chowdhury et al., 2015; Varlan and Sansen, 1996), taste (Fang et al., 2007), ageing (Bakr et al., 2016) and bruising (Jackson and Harker, 2000).

Most EIS studies were carried out using bench-top impedance analysers, which combine the possibility of using a wide range of test frequencies and different electrode configuration with a high measurement accuracy. The limitations of such tools in terms of price and size has led to the development of low-cost microcontroller-based systems, that are suitable as portable EIS analysers (Al-Ali et al., 2017; Chowdhury et al., 2018; Simic, 2013). The drawback of these portable systems, in terms of lower accuracy and quality of the output signal, compared with bench-top instruments, is generally compensated by the lower costs and higher portability, which constitute a major advantage for on-field applications.

Regardless of the system, the measured impedance spectra has to be fitted with an equivalent circuit model to provide information about the properties and behaviour of the sample under study (Grossi and Riccò, 2017), and that provides information about the electrochemical processes in the tissues (Grimes and Martinsen, 2000). The flow of current resulting from an external electrical field has been described by several models, which can be divided in two main categories: integer-order models and fractional order models. Integer-order models represent the flow of current into intra and extra-cellular fluids with resistances, whereas cell membranes are modelled through capacitors (Hayden et al., 1969; Zhang and Willison, 1991). On the other hand, fractional-order models use the resistances to simulate different tissues among the plant cells, while the capacitors are replaced with constant phase elements (CPE), in order to model non-ideal capacitor behaviour. Here, the most widely known fractional-order methods are the single and double dispersion Cole impedance models (Cole, 1940). Nevertheless, equivalent circuit models have drawbacks. The former ones, while explaining the underlying physiological mechanisms generating the electrical response of the fruit, are not able to achieve a good fit of the impedance data without adding a high degree of complexity to the circuit, which in turns complicates the interpretation of the results. The latter ones, in contrast, have extremely good fit to the bio-impedance curves, but lack physiological interpretation of the electrical response of the samples under test (Freeborn, 2013). Neither class of models takes into account the interaction at the interface between the electrodes and the sample surface, which has a non-negligible influence on the final impedance output. An equivalent circuit model that can properly fit the bio-impedance of the fruit while considering the electrode-fruit interface and explaining physiological changes in the sample, is necessary.

In the present work, EIS data of apples and bananas were acquired over a 13 d period to study the effect of ageing. The experimental data was acquired in the 100 Hz - 85 kHz frequency range using a microcontroller-based EIS system. First, the bio-impedance data were evaluated in terms of their absolute evolution, as well as in terms of the influence of the applied frequencies. Secondly, the measurements were fitted with a proposed equivalent circuit, that presents a Warburg element to model the electrode-fruit interface and a simple circuit consisting of only four components (one Warburg element, two resistors and one capacitor). This model achieved satisfactory fitting results, (fit Root Mean Squared Error of 2.66% and 2.82% for apple and banana, respectively), in line with the results obtained by the models already in use elsewhere.

2. Materials and methods

2.1. Bio-impedance measurement

Nine apples and nine bananas were purchased from a local dealer and kept at room temperature for 13 d. Bio-impedance data were measured every 24 h. An AD5933 evaluation board (Analog Devices, Norwood, MA, USA) was used to extract the magnitude and phase angle data in the frequency range between 100 Hz and 85 kHz. The spectrum was chosen according to the literature on the application of EIS on fruit and to the evaluation board factory limitations, which can provide the excitation voltage in the 5 to 100 kHz frequency range. The upper limit was set to 85 kHz to avoid possible measurement errors at to the instrument limit. Although its upper limit cannot be expanded, with additional circuitry the evaluation board can reach lower frequencies. Indeed, the frequency input between 100 Hz and 8100 Hz was obtained by means of an AFG-3031 function generator (Good Will Instrument Co., Taiwan), which provided a 2 MHz signal, while the higher frequency range between 5 kHz and 85 kHz was obtained using the 16 MHz evaluation board internal clock. The frequency range used to analyse the data was optimized by excluding the first noisy frequencies and the overlapping points, resulting in 123 frequency points between 400 Hz and 85 kHz. The fruit samples were excited with a 1 V p-p sinusoidal signal, with a DC bias of 0.76 V, automatically applied by the AD5933 impedance converter chip on the evaluation board. The system accuracy was verified by calibrating the system using capacitors in the 10 nF–100 nF range, resulting in no significant difference between the theoretically calculated impedances and the measurement. The electrical contact with the samples was established by means of pre-gelled ECG (Ag/AgCl) electrodes (Fiab F3001 ECG). A total of 4 electrodes was placed on the fruit at a 3 cm distance between each other for the banana (Fig. 1A) and with equal spacing along the maximum radius for the apple (Fig. 1B). The electrodes were maintained on the fruit surface during the experiment duration, providing more reliable measurements. As a result, electrode spacing, impedance and degradation rates remained constant during the experiment duration. Each fruit impedance spectrum was then obtained by averaging the measurements, carried out using a 2 electrodes-configuration, performed with different couples of electrodes. Four for the banana (1–2, 3–4, 1–4 and 2–3) and

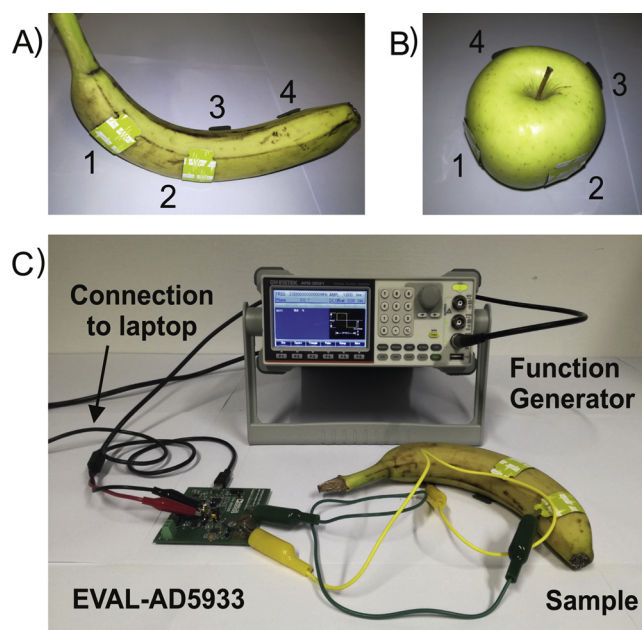


Fig. 1. Electrode placement on banana (A) and apple (B) and experimental setup of the impedance measurement with AD5933 and function generator (C).

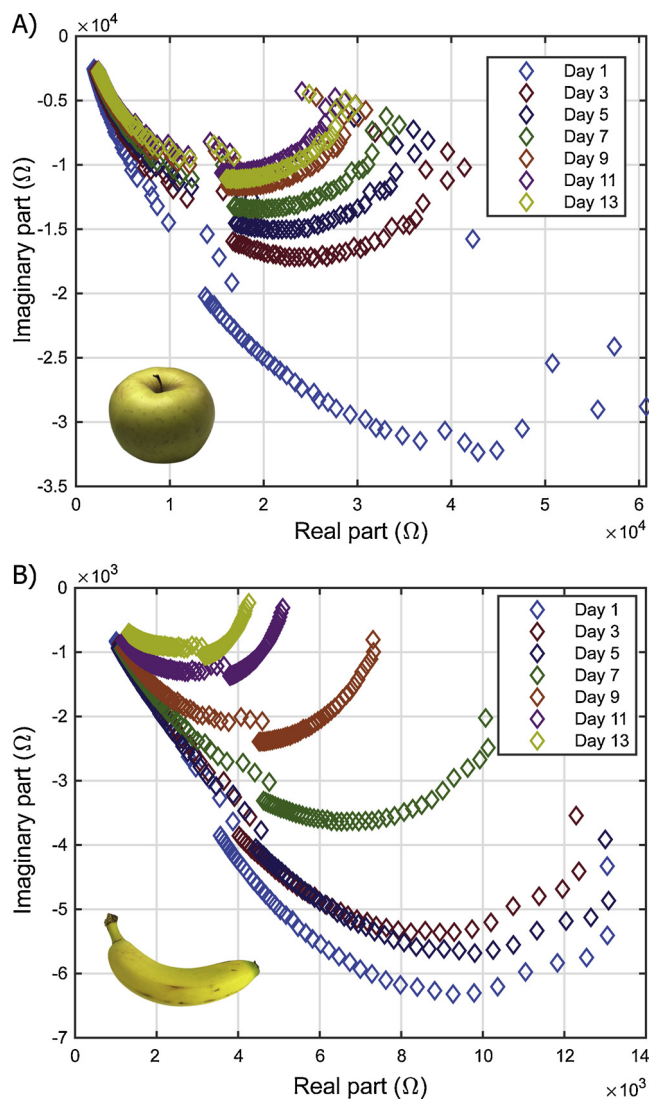


Fig. 2. Nyquist plot of the average impedance of nine fruit measured every 2 days from day 1 to day 13 for apple (A) and banana (B).

three for the apple (1–3, 2–4 and 1,2), respectively, with the objective of obtaining an output representative for the entire fruit (Fig. 1).

2.2. Software and data analysis

A laptop computer coupled with a custom-made LabVIEW program (Version 2016, National Instruments, Austin, Texas) was used to acquire the impedance data in terms of magnitude and phase angle. Data were then processed using Matlab R2017b (The MathWorks Inc., Natick, MA, United States). The impedance data were fitted with the equivalent circuits using Matlab Zfit script (by Jean-Luc Dellis, version 1.2.0.0, retrieved February 2018). A schematic of the experimental setup is displayed in Fig. 1C.

3. Results and discussion

3.1. Impedance and frequency

The Nyquist plot for banana and apples over 13 d in shown in Fig. 2. The apple results show an average impedance an order of magnitude larger compared to the banana. The former values lay in a range between 18 kΩ on day 1 and 12 kΩ on day 13, while the latter ranges between 4 kΩ in the first day of acquisition and ends at around 2 kΩ in the last day. Despite the similar decreasing trend, the evolution of the impedance also highlights a remarkable difference among the two fruit. The apple had the biggest impedance reduction between the first two days of acquisitions, after which the impedance gradually decreased and stabilized. In contrast, the banana shows a first steady phase during the first 5 d, with no consistent variation of the impedance. The differences in the physiological characteristics between the two fruit highlight the potential of this technique in tracking the ripening of deeply different fruit using a micro-controller-based system. Furthermore, the observed decrease over time of the impedance are consistent with the literature on such fruit, based on bench-top and microcontroller-based instruments (Bakr et al., 2016; Chowdhury et al., 2015; Ibba et al., 2018; Mohapatra et al., 2017; Watanabe et al., 2018). To investigate the influence of the frequency on the impedance change of the fruit, principal component analysis was applied using a reduced dataset: first, each fruit impedance spectrum for each day was averaged to obtain the mean daily impedance; secondly, the 123 frequency points (in the 400 Hz - 85 kHz range) of each spectrum were grouped in 11 frequency clusters and averaged, obtaining a final dataset of 11 impedances for each of the considered days. The results (Fig. 3)

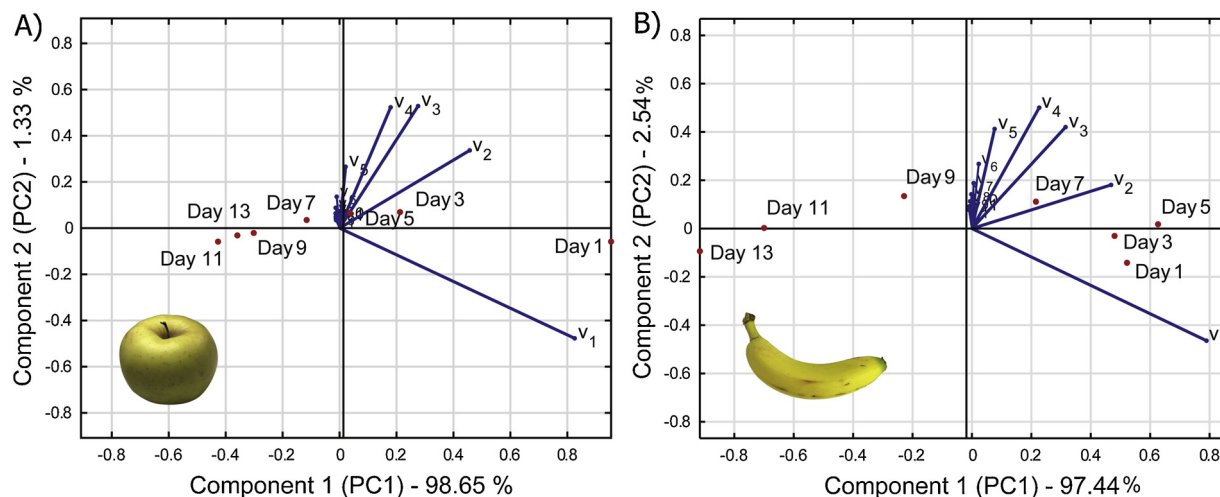


Fig. 3. Bi-plot of principal component analysis restricted to the first two components, which explain the 99.98% of the variance for both apple (A) and Banana (B). The red dots indicate the mean impedance values for the days, while the blue lines indicate each frequency cluster. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

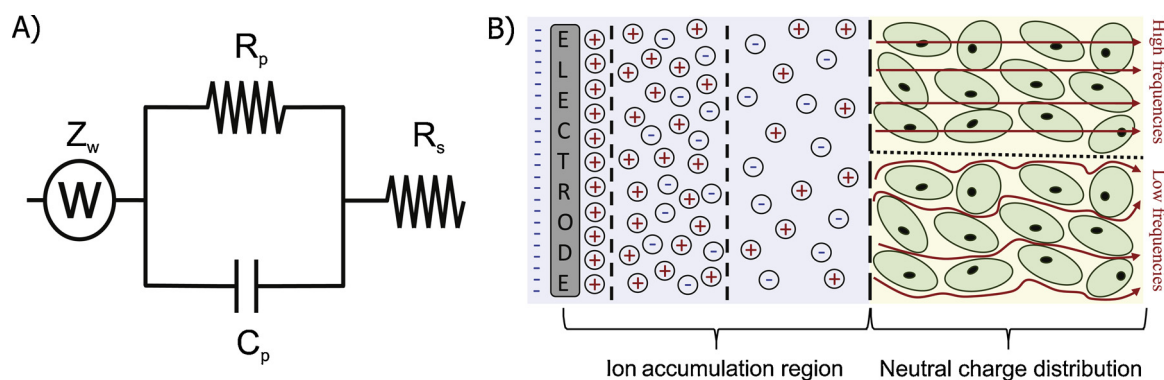


Fig. 4. Proposed equivalent circuit model (A) and general current flow in fruit tissues (B). The left side represents the ion accumulation region at the sensor fruit interface, while the right side depicts the current flow in the neutral charge distributed region, at high and low frequencies.

highlighted a strong correlation of the lowest frequency points, corresponding to the first 4 frequency clusters (400 Hz to 6 kHz), with the impedance change occurred during the study for both fruit.

Furthermore, the first two principal components explained the majority of the variance (99.98% for both fruit), but with different behaviour. Along the first principal component (PC1) it is possible to observe a clear separation among each day of acquisition, that follows the general trend observed in the Nyquist plots. The second principal component (PC2), although it does not have a great impact on the apple impedance, explains the apparent stable behaviour that occurs during the first 5 d of sampling of the banana. In Fig. 3B it is clear how the PC2, representing just the 2.54% of the variance, contributes to a separation of the above-mentioned days that the PC1 alone would not be able to achieve, albeit contributing to the 97.44% of the total variance.

3.2. Equivalent circuit

The average daily impedance output for each fruit was fitted with the equivalent circuit in Fig. 4A.

The circuit was designed with the purpose of providing the best fit of the impedance data while minimizing its complexity (i.e. the number of components), while simplifying the biological explanation of the impedance output. The Warburg element was added to the circuit to model the interface between the fruit surface and the electrode. The Warburg impedance can be associated to the mass diffusion process occurring at the interface between an electrode and an electrolyte (Warburg, 1899), specifically the diffusion of ionic species towards the electrode (as shown in the ion accumulation region, in Fig. 4B). In this way, the electrode-fruit interface can be compared to an electrical double layer, which contains counter-ions (with opposite charge than the electrode) and co-ions (with the same charge as the electrode). This electric double layer is characterized by a finite thickness, which represents the limit of the electrostatic influence of the electrode in its vicinity and strongly depends on the concentration and size of the ionic species present in the medium (Bohinc et al., 2001). Furthermore, it is well known that in AC measurements a high frequency sweep results in a reduction of the ions diffusion, together with an ion concentration gradient increase at the interface (Yufera et al., 2011). However, considering the frequency range used in this experiment and the influence of the low frequency points on the impedance change, it can be assumed that the process of ion diffusion reduction at high frequencies is negligible. Therefore, the Warburg impedance has to be considered a central element in the equivalent circuit. The remaining elements were used in accordance to the existing state of the art (Cole, 1940; Hayden et al., 1969; Zhang and Willison, 1991). The capacitor (C_p) is added to model the dielectric interface behaviour of the cellular membrane, while the resistors are typically used to model the current flow in liquid mediums, which in case of the fruit are represented by extra and intracellular fluids. In particular, given the typical frequency response of an

ideal capacitor (an open circuit at low frequencies, a short circuit at high frequencies), it is possible to account for two different phenomena observed in cell membranes. At low frequencies, the cell membrane, having a high impedance, acts as an open circuit forcing the current to pass around the cells in the extra-cellular fluid, modelled by the parallel resistor (R_p). In this case, the impedance output, neglecting the Warburg impedance, will be composed mainly by the series of R_p and R_s ($R_s + R_p$). However, at high frequencies the cell membrane acts as a short circuit, letting the current flow everywhere in the intracellular fluids, modelled by the series resistor (R_s), that present a higher concentration of electrolytes compared to the extracellular-one. Hence, the resistance value will be composed by the parallel of R_s and R_p ($R_s // R_p$), in which R_s is the dominant component, lowering the general impedance output (Watanabe et al., 2018).

3.3. Model fitting

The experimental data were fitted with both the proposed equivalent circuit model (Fig. 5A) and with the models already presented in literature, to validate our method. The results were first evaluated in terms of Root Mean Square Error (RMSE) of the estimated data compared to the real one. Secondly, changes of the circuit parameters during the fitting of different days were assessed to correlate it with a physiological behavior of the fruit during ripening. Fig. 5 shows the fit and its changes during two weeks of storage for apples (Fig. 5A) and bananas (Fig. 5B), using the equivalent circuit model proposed in this paper.

The fitting procedure was carried out following three main steps. First, the best starting parameters (Y_0 , B , C_p , R_p and R_s) for the fit at day 1 were selected to iterate different combinations of starting values of the circuit components, and selecting those giving the best fit, as identified by the MatLab Zfit script. Subsequently, the best circuit parameters used for the best fit for each day were used as a starting point for fitting the following acquisition day curve. Finally, the best fit for each day was extracted, along with the best obtained circuit parameters, in order to evaluate the output in terms of fit quality. Moreover, the RMSE of this equivalent circuit was evaluated and compared with the ones of other models presented in literature for the application of EIS to fruit. Specifically, the impedance output was tested on the classical integer-order models (Hayden et al., 1969; Zhang and Willison, 1991) and on the most recently applied and better performing fractional-order models, namely the single and double dispersion models (Cole, 1940). Fig. 6 highlights the difference in fit quality between the models for apples and banana.

As expected, the integer order models yield a poor fit quality compared with other models. In particular, the Hayden model had the worst results (RMSE > 15% for apples and bananas), both in its classical and in its simplified version, as confirmed by the work of Stout et al. (1987). An improvement in the fit was achieved with the Zhang model, widely

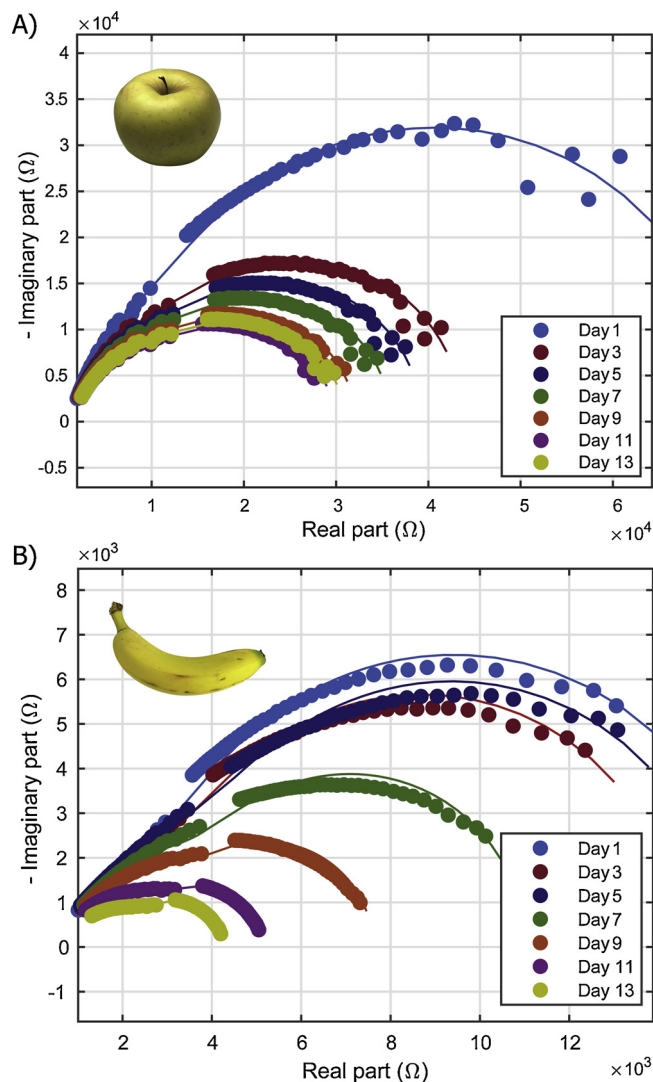
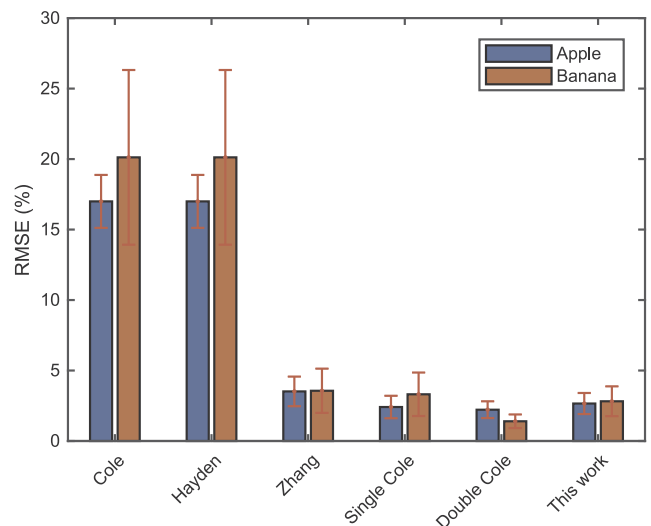


Fig. 5. Proposed equivalent circuit model fitting for apple (A) and banana (B). The dots represent the experimental results, while the lines depict the optimized curve for each day.

used in bio-impedance, that has a RMSE around 3.5% for both fruit, but it is characterized by more parameters to correlate with the fruit status. On the other hand, the fractional order models gave the best results in terms of fit quality, confirming the results found in literature (AboBakr et al., 2017), especially for the double Cole, that shows a RMSE as low as 2.22% for the apple and 1.40% for the banana. However, as discussed above, such fit is not fully supported by a good biological explanation of the underlying mechanisms. Finally, the model proposed in this work shows a good fit, with comparable or better results than the fractional order models, especially in the case of the single Cole, with a RMSE of 2.66% and 2.82% for apples and bananas, respectively. This confirms that the electrode-fruit interface, modelled with a Warburg element, contributes to a great part of the impedance output, especially in the low range of frequency used in this work. This aspect is also strongly related to the simplification of the equivalent circuit, changes of parameters being better correlated to a quality change in the fruit, as it will be discussed in the next section.

3.4. Circuit parameters evolution

The application of electrical impedance spectroscopy on a fruit, especially while monitoring its physiological changes over time,



Model Name	Equivalent circuit	Apple		Banana	
		RMSE (Ω)	RMSE (%)	RMSE (Ω)	RMSE (%)
Cole		2370	16.99	782.7	20.12
Hayden		2370	16.99	782.7	20.12
Zhang		498.6	3.52	141.5	3.56
Single Cole		343.2	2.41	128.6	3.31
Double Cole		313.9	2.22	54.3	1.40
This Work		376.5	2.66	110.8	2.82

Fig. 6. Fitting quality comparison. (A) Evaluation of the percentage RMSE and standard deviation (SD) among different equivalent circuit models for apples (blue) and bananas (red). (B) Equivalent circuit: absolute value and percentage value of RMSE for considered equivalent circuit models. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

requires the correlation of the measured impedance output with the inherent physiological mechanisms occurring during its ripening. In particular, next to the evaluation of the impedance progression and its dependence on frequency, it is important to assess changes of the overall circuit components (Fig. 7A), to better correlate their behaviour with fruit ageing. In Fig. 7B–F, the average trend of each circuit components over 13 d is displayed for apples and bananas.

The main quality factor affecting the fruit impedance variation during storage is its water content and the relative concentration of charged particles in it, as confirmed for apples (Watanabe et al., 2018),

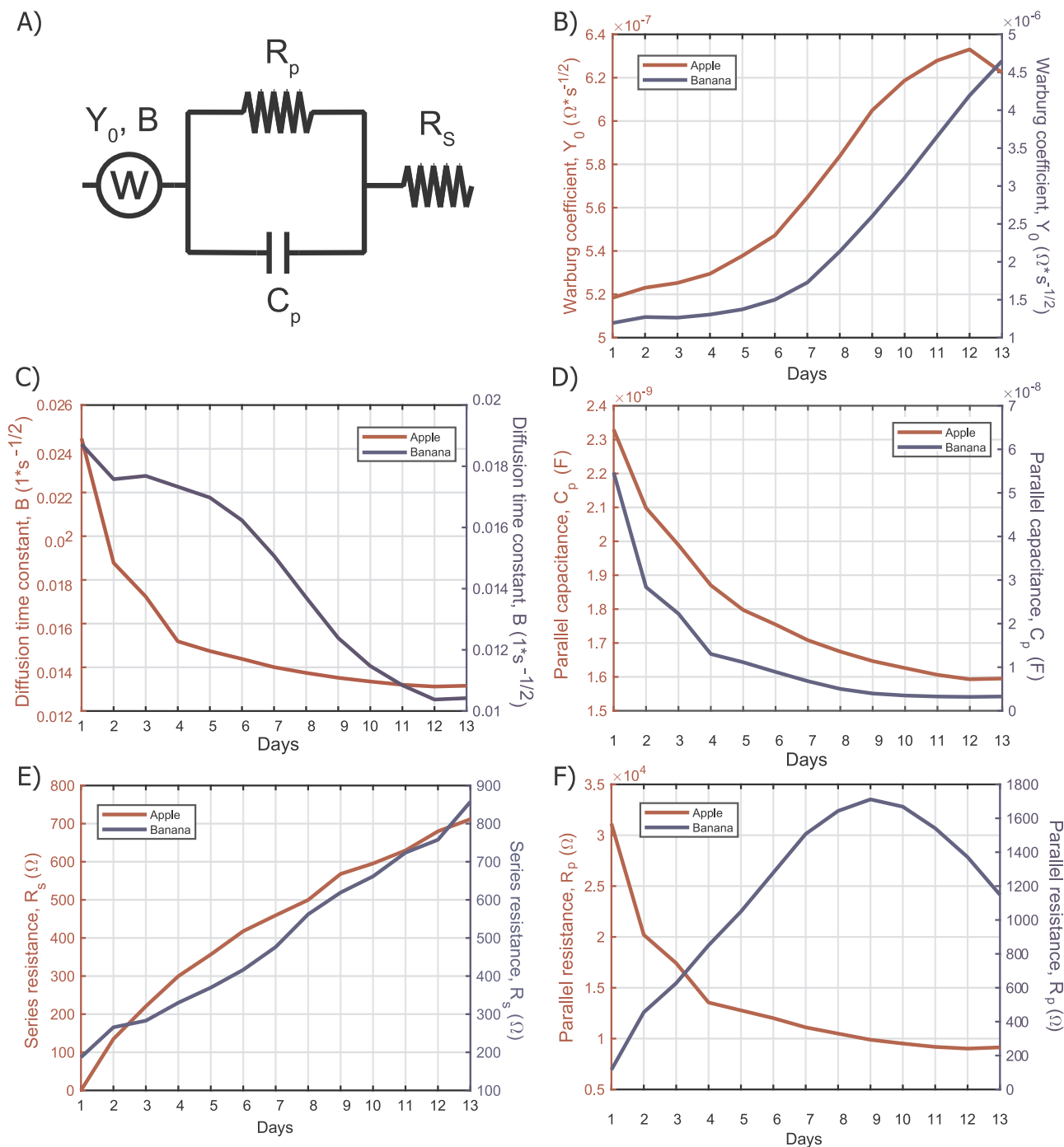


Fig. 7. Proposed equivalent circuit (A) and its parameters evolution for Warburg coefficient Y_0 (B), diffusion time constant B (C), parallel capacitance C_p (D), series resistance R_s (E) and parallel resistance R_p (F) for the average of 9 apples and 9 bananas.

and bananas (Mohapatra et al., 2017). From an electrical point of view, the current flows in liquid mediums due to the presence of charged particles, or ions, that are mainly represented by minerals in fruit. During storage, absolute losses of minerals are impossible, while an absolute mineral content increase is possible as a concentration effect, consequently to transpiration losses (DeEll and Prange, 1993). Transpiration is the evaporation of water from the plant tissues and its rate is determined by both internal factors, such as morphological characteristics, maturity stage and surface-to-volume ratio, and environmental factors, such as temperature, relative humidity, and atmospheric pressure (Kader and Yahia, 2011). Therefore, we can consider the water loss via transpiration, and the consequent increase of absolute

ion concentration, as the main factor involved in the change of the equivalent circuit parameters.

In Fig. 7B and C the Warburg coefficients are displayed. In particular, the diffusion admittance, (Y_0) and the diffusion time constant (B) parameters respectively, are related to changes of the Warburg impedance, using the following Eq. (1):

$$Z_w = \frac{1}{Y_0} \tan h(Bj\omega) \tag{1}$$

Where ω , which is the angular frequency, can be considered negligible, due to the fact that the same frequency range was considered for each daily average impedance, for both fruit. The values of both Y_0 and B

change over time. The admittance value Y_0 increases while the general impedance decreases, and the diffusion time constant value B decreases. This is probably due to a higher concentration of charged ions in the medium, that increases the general conductivity, influencing Y_0 , and decreases the diffusion layer thickness at the fruit-electrode interface, hence reducing the diffusion time constant B .

As discussed in Section 3.4, the circuit elements R_s , R_p and C_p model the current flow through intra and extra-cellular fluids and therefore are useful to monitor the fruit water status. An additional physiological factor that can contribute to the explanation of the changes of such elements, especially over time, is represented by senescence. Senescence is a genetically controlled process, triggered at the onset of ripening, which produces a sequential deterioration of the cellular structure (Atkinson et al., 1980), with a consequent electrolyte leakage in the extracellular fluids. There is a gradual reduction of the C_p values in for both fruit (Fig. 7D). This can be explained with the progressive deterioration of the cell membrane due to senescence. The intracellular resistance (mainly represented by the series resistor R_s) values, shown in Fig. 7E, depicts an increasing trend for both fruit, with similar start and end points. Accumulation of water by the cells, with consequent dilution of the ionic concentration in the medium, with a consequent drop in conductivity is likely associated with a homeostatic process that takes place to maintain the cell turgor, weakened by the gradual deterioration of the cell membrane. Finally, the most interesting curves are those connected to the extracellular fluid (represented by the series of the resistors, $R_p + R_s$). Each fruit has very different trends (Fig. 7F). The apple shows a first decrease in resistance at the beginning of storage, consequent to a first transpiration peak, which tends to stabilize at day 4. This results in an initial concentration of electrolytes in the medium, that lowers the resistance. In contrast, in bananas the R_p value steadily increased from day 1 to day 9, and then decreased, probably because during transpiration, water moves from the pulp to the skin, resulting in a lower conductivity of the extracellular fluid. Furthermore, changes towards later time points can be attributed to the onset of the final senescence stage, which cause the cell to deteriorate to a level where the leakage of intracellular fluids, higher in ion concentration, cannot be considered negligible. Behaviour of the fruit was also confirmed by a visual evaluation in which the apples appeared to be in good condition while the bananas were completely black. The circuit parameters helps to better understand the changing impedance described in Section 3.1. The conductivity change in the extracellular environment, represented by R_p , is the factor influencing the most the impedance change in terms of absolute value and behaviour. It reproduces the onset of a relatively steady decreasing state for the apples and the reaching of the final senescence state for the banana. These results are helpful to give a better insight of the processes occurring during ripening and senescence. However, to transfer this technique from a laboratory to a field application will require the development of new portable and low-cost acquisition systems. The correlation with other fruit quality parameters (e.g. sugars and acids content, texture) also should be addressed using predictive models.

4. Conclusions

A microcontroller-based EIS system was used to extract the impedance data of apples and bananas in a frequency range from 100 Hz to 85 kHz, and to monitor the progress of the ripening during 13 days at room temperature. The impedance output was followed with a focus on the influence of the applied frequencies on the changes. Subsequently, the impedance output was fitted with integer-order and fractional-order equivalent circuit models found in literature, and the results were compared in terms of goodness of fit with the proposed equivalent circuit. Lastly, the equivalent circuit components were evaluated during the fruit ripening to correlate and to monitor the physiological behaviour of the samples under test. A relationship between the fruit ripening and the impedance was found. In particular, the system was

capable of tracking the fruit maturation curves, highlighting a strong correlation of the low frequencies of the spectrum with the impedance change for both fruit. The proposed equivalent circuit model showed a comparable goodness of fit with the models found in literature and was able to fit the variation of the impedance curves during the ripening. Furthermore, both the flow of current in the fruit and the mechanisms occurring at the electrode-electrolyte interface could be modelled, which has not been considered in previous studies. The addition of the Warburg element to the circuit was essential to address this problem, as the ion movement occurring towards the electrode surface could be modelled. That it has an impact on the impedance measurement, improves the state of the art on the application of this technique on fruit. Finally, the model circuit parameters evolution was in line with the physiological changes occurring during the fruit ripening and senescence, especially in terms of cell wall degradation, water movement and electrolyte concentrations. Such results, giving a better insight on the electrical properties of apples and bananas, can be employed as a starting point for future studies on the correlation of the chemical composition and the early detection of diseases. In conclusion, we believe that this cost effective and simple method, coupled with a good biologically explainable equivalent circuit model fitting and, in future, with predictive models for fruit quality, will represent an excellent low-cost alternative for the non-destructive and in-line evaluation of fruit quality.

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