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SYSTEM DEVELOPMENT FOR MONITORING THE PRODUCTION PROCESS OF FREEZE-DRIED SAMPLES: A SIMPLE AND LOW-COST APPROACH

Article Highlights

- Accurate and precise data on the freeze-drying process were obtained using low-cost sensors
- The use of correction blank curves significantly reduced the sample's mass measurement errors
- It was identified that the PI controller is suitable for the production process of lyophilized samples
- The system developed could also be used to acquire data on the freezing process

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Abstract

The data acquisition from the freeze-drying process is important for obtaining freeze-dried samples with the desired final moisture content under various operating conditions. The current study extensively presents a simple and low-cost methodology for implementing a data acquisition system in a laboratory-scale freeze dryer. The results showed that higher drying temperatures (40 °C) increased the errors involved in measuring the mass of material; nevertheless, the application of correction blank curves statistically significantly reduced those errors. In general, the system developed provided precise and accurate measurements of the temporal changes in the sample mass and temperature, and chamber pressure variations, allowing monitoring of the production process of freeze-dried samples with low final moisture contents.

Keywords: lyophilization, drying, freezing, heating temperature, Arduino, avocado.

SCIENTIFIC PAPER

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Freeze-drying is a dehydration process whereby the amount of water in a material is reduced by freezing the liquid fraction, followed by sublimation of the ice. Low temperatures reduce the possibility of food degradation, so lyophilized products have better nutritional quality and sensory characteristics [1,2].

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However, the wider application of the technique is limited by slow drying rates and high operating costs in terms of the energy required for freezing and vacuum. Consequently, freeze-drying is restricted to products with higher added value [3–5]. To reduce the drying time, heat can be supplied to the sample; nevertheless, the heat flux must not be excessive; otherwise, the material can be harmed [6–8].

To study the effect of operational conditions on the final product quality, samples of the material are freeze-dried under different conditions in small-size pilot plants or even lab-scale freeze-dryers [9–11]. The final moisture content of the freeze-dried material is an important parameter that affects product quality and energy consumption. Thus, it is important to monitor the

sublimation rate of the material during the freeze-drying process [10]. To measure this data, many technics have been utilized, such as discontinuous measurement of the sample moisture [12], determination of freeze-drying rate by artificial neural networks [13], thermal flux sensors [14], Tunable Diode Laser Absorption Spectroscopy (TDLAS) [15], Manometric Temperature Measurement (MTM) [16] and Pressure Rise Analysis (PRA) [17], and real-time measurement of sample weight throughout the drying process [10]. In general, most of the experimental data shown in the literature were measured by this first method, which is not advantageous, once the number of experimental points is limited, and the interruptions of the process may lead to long operational times [9,10,18–21]. On the other hand, this last method is reported by some authors as accurate and reliable [13,22]. In addition, it is possible to simultaneously determine the mass and temperature of the sample, which is useful for identifying the transition from the primary to the secondary drying stage [23].

However, commercial freeze dryers with integrated real-time data acquisition systems are rare and often expensive, which leads to the need to adapt conventional freeze dryers. In this context, a few studies in the literature show the adaptation process for research purposes, final moisture control, and sample production. Pikal, Shah, Senior, and Lang [24] described the direct experimental determination of the resistance of the lyophilized product using a suspended microbalance. The system was successfully applied to monitor the drying process of several aqueous solutions. Carullo and Vallan [9] and Vallan [23] describe a system for monitoring the mass and temperature of samples during the lyophilization process. In this system, heat was provided to the sample by conduction. To reduce the mass measurement errors induced by the heat supply, a mechanical lifting system had to be used to raise and release the vials when the mass measurement had to be carried out. Although this system showed good results, the lifting process may affect the lyophilization speed and increase the complexity and costs of the system. In the same way, Roth, Winter, and Lee [25] also used a mobile microbalance. However, in this method, the heat supply was interrupted during the mass measurements. Tribuzi and Laurindo [10] showed a freeze-dryer data acquisition system where a mechanical lifting apparatus was not required, and heat was also provided to the sample by conduction. The authors explored the stability of the mass measurements in blank experiments and the drying rate and temperature data of the freeze-drying process of banana slices. Nevertheless, for simplicity of the mass measurement system, the supply of heat by thermal radiation is preferable once scales with floating sample holders can be employed [23]. Kirmaci *et al.* [11] and Menlik *et al.* [13] showed only some details of the data acquisition systems utilized in the freeze-drying process of apples and strawberries, with heat supply by thermal radiation, but the methodology of adaptation of the freeze-dryers was not reported. Xiang *et al.* [26] constructed a custom microbalance to better control and measure sample temperature and chamber pressure. They observed that the microbalance was useful for studying the sublimation rate during lyophilization. Fissore *et al.* [27] and Pisano [21] provided reviews of the different approaches to using microbalances to study the lyophilization process. They concluded that the system could be useful, mainly to determine the mass transfer resistance of the sample.

In general, most of the systems cited use high-cost sensors, and several important details were not reported in the papers, such as the main physical structure adaptations necessary, electrical/electronic diagrams, and signal noise reduction procedures. It is highlighted by Fissore *et al.* [27] that depending on the operational conditions, process fluctuations, vibrations, gas flows, and temperature gradients can affect the measurement performance of different sensors. In addition, most of those papers did not analyze the main performance indicators of measurement systems, such as accuracy, precision, and stability. In this context, the need for detailed methodologies to correctly develop a system for monitoring the production process of freeze-drying samples is observed, mainly when the heat is supplied to the sample holder by thermal radiation. It is important to highlight that, from a practical point of view; the incorrect measurement of the drying kinetics data can lead to a misleading estimation of the drying time required for obtaining the desired final moisture content of the freeze-dried product. Thus, a detailed methodology for developing a system for accurate data acquisition of the chamber pressure and temperature and sample mass and temperature variation through the freeze-drying process stands out. In particular, the high accuracy of the sample's mass measurement is an important factor since, for some materials, the variation throughout the drying process is very small.

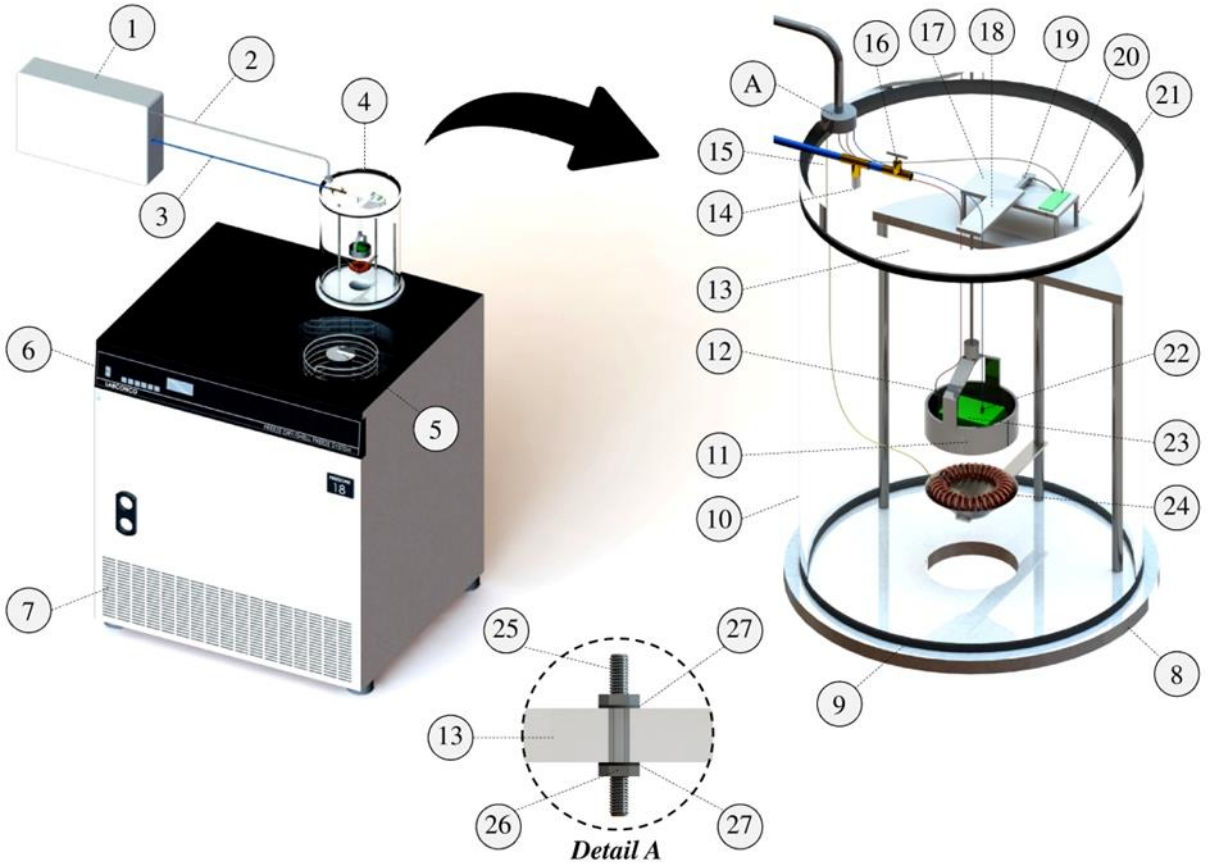
Thus, this work aimed to describe in detail the adaptation process of a laboratory-scale freeze dryer by introducing a data acquisition system for monitoring the material's mass and temperature, and chamber pressure, during the production process of freeze-dried samples. The acquired data were used to control the final moisture content of the produced sample. The system was built using low-cost sensors, actuators, and microprocessors, developing a more accessible and

reproducible methodology. The performance indicators of the mass measurements in blank experiments were determined for different operational conditions. As a test case, the system's performance was also evaluated in drying avocado (*Persea americana*) pulp, with tray heating temperature at 40 °C and without temperature control. The results were used to determine energy indexes and transition points during the drying process. The system's reliability was analyzed by comparing the mass of material determined by the data acquisition system, before and after the drying process, with the data provided by an external analytical scale.

MATERIALS AND METHODS

System development for freeze-drying data acquisition

The studies were performed using a laboratory-scale freeze dryer (Labconco®, FreeZone, console 6L), as shown in Figure 1. The basic components of the system were: data acquisition system (1), drying chamber (4), vapor condensation chamber (5), control panel (6), and freeze dryer internal components (7).



Captions

1 – Data acquisition system:
  a – Microprocessor (Arduino®, Mega, 2560R3)
  b – Pressure transducer (Freescale®, MPX5500DP)
  c – Pressure, temperature, and humidity sensor (Bosch®, BME280)
2 – Data transfer cable and electrical commands
3 – Flexible pneumatic hose
4 – Cylindrical acrylic chamber
5 – Vapor condensation chamber
  a – Temperature sensor (Maxim Integrated®, DS18B20)
6 – Freeze dryer control panel
7 – Freeze dryer internal components:
  a – Vacuum pump (JB Industries®, DV 142N 250)
  b – Radiator Heat Exchanger
8 – Steel support of the cylindrical chamber
9 – Rubber seal of the cylindrical acrylic chamber
10 – Acrylic wall of the cylindrical chamber
11 – Aluminum sample holder tray
12 – Temperature sensor (Maxim Integrated®, DS18B20)
13 – Acrylic chamber cover
14 – Pressure tap point
15 – Temperature sensor (Maxim Integrated®, DS18B20)
16 – Acrylic support of load cell
17 – Pressure relief valve
18 – Acrylic extension of load cell
19 – Load cell (Zhipu®)
20 – Voltage amplifier (HX711, 24 Bits)
21 – Helical springs
22 – Temperature sensor (Maxim Integrated®, DS18B20)
23 – Avocado sample
24 – Stainless steel resistor (25W)
25 – Steel screw
26 – Steel screw nut
27 – Rubber o-ring

Figure 1. Experimental system for data acquisition of the freeze-drying processes.

The vacuum pump (7a) specification was 5 CFM, two stages, a final vacuum of 3.2 kPa, and a flow rate of 8.52 m^3/h. The drying environment consisted of a cylindrical acrylic chamber (4). The junction between the cylinder and the cover was hermetically sealed with a rubber ring (9). To standardize the experiments and reduce random errors, such as ambient heat transfer by thermal radiation to the sample, all the assays were performed in a climatized environment (ambient temperature of approximately 22 °C), close to the sunlight, under an artificial light source (fluorescent lamp of 32 W). To obtain data on the freeze-drying process, the drying chamber (4) and drying chamber cover (13) were modified by the attachment of a mass measurement system (11, 17, 18, 19, 20, and 21), a heating system (24), and temperature (5a, 12, 15 and 22) and pressure (14) sensors.

The mass was measured using a load cell (19) with an operating temperature range from -20 to 60 °C, a maximum load of 200 g, and a precision of ±0.04 g. The load cell (18) was attached to the acrylic support (17) by two screws. This support was coupled to another one of larger proportions using four helical springs (21) to minimize the effect of vibrations on load cell measurements. The load cell had an extension (18), attached by two screws, that was positioned centrally in the drying chamber. An aluminum sample holder tray (11) was suspended from the load cell extension (18) by a steel cable. In this way, the load cell was sensitive to changes in the mass of the sample holder tray (11). The low-strength signals produced by the load cell were amplified by a voltage amplifier (20) and transmitted by cables (2) to a microprocessor (1a). The electrical diagram in Figure S1 (Supplementary Material) details the wire connections between these devices. The load cell was calibrated using different weights from 1 g to 200 g. A touch button (Figure S1) was added to the system to tare the balance before each freeze-drying procedure.

The tray was heated from underneath by irradiation from a 25 W resistor (24), powered by a 12 v power source, detailed in Figure S1. The temperature of the tray was measured using a sensor (12) with a range from -55 °C to 125 °C and a precision of ±0.5 °C. The temperature data of the tray was also transmitted through cables (2) to the microprocessor (1a). These data were used by the microprocessor to act on a solid-state relay (Figure S1) connected to the heating resistor (23) and feedback-control the temperature of the tray using an on-off control. The routine for data reading, control, and writing was developed using Arduino® IDE 1.8.5 software and can be accessed in section S.1 (Supplementary Material). The distance between the resistor and the tray was 50 mm. The temperatures of the sample (22), the chamber ambient (15), and the condenser surface (5a) were measured by sensors of the same model and also transmitted to the microprocessor. Note that flexible cables were used for the electrical wiring of the temperature sensors (12 and 22) placed on the sample holder tray (11), which were attached to the load cell extension. This configuration was adopted to reduce the influence of the movement of the cables on load cell measurements. The temperature sensors were calibrated using a calibrator block (Tchne®, Model DB-35L) in the temperature range employed in the experiments.

A t-connector was attached to the drying chamber cover (13) so that the pressure (14) inside the chamber could be accessed. Thus, one outlet of the t-connector leads to a needle valve (16) for removing the vacuum. The other outlet was connected (3) (using a ¼" poly-flux hose) to a pressure sensor (1b), with an operating range from 0 kPa to 500 kPa and an accuracy of 2.5%, for measuring the internal pressure of the vessel. The pressure data was also transmitted to the microprocessor. Figure S1 details the wiring connections between these devices. The pressure sensor was calibrated with an air vacuum/pump station (Cole-Parmer®, Model 470-5942).

A pressure, temperature, and humidity sensor (1c) with the accuracies of ±3% (humidity), ±0.5 °C (temperature), and ±0.1 kPa (pressure) was used to monitor the environmental conditions outside the chamber during the freeze-drying processes.

The wire connections of the load cell, temperature sensors, and electrical resistance were made with stainless steel screws (25) through the acrylic chamber cover (13), sealed with o-rings (27), as shown in Detail A of Figure 1. This type of connection was necessary to prevent vacuum leakage. The sensor data were acquired by the microprocessor (1a) and were transmitted to a computer (Intel® Core™ i7-930 processor, 4 GB RAM) through a USB cable (baud rate of 9600). The data were recorded in real-time, using a Microsoft® Excel spreadsheet. The communication between the microcontroller and the Excel software was performed using the PLX-DAQ add-in (Parallax Inc.).

It's important to highlight that the system developed in this study proposes the use of a microcontroller based on the Arduino® platform due to its low cost, high processing velocity (16 MHz), and relatively good analog input channel resolution (10 Bits), when compared to other commercial products available or proposed by previous works [11,25,28]. Arduino® is open-source hardware with easy installation, control, and operation that does not require

extensive technical support [29–32]. In addition, all sensors used are easily accessible and of low cost, which facilitates the use of the methodology described in this work.

Evaluation of the online mass measurement system

To verify the reliability of the obtaining methodology of mass measurements in the drying chamber, over 80,000 blank data were acquired before and after the freeze-drying procedures for 20 h. The tests were performed in duplicate, without adding a test specimen (avocado), with a heating temperature of 40 °C and without temperature control, to observe the effect of temperature on the stability of the measurements. The vacuum pressure and the condenser temperature were approximately 0.2 kPa and -50 °C, respectively.

The data were analyzed using the main performance indicators of measurements, such as precision and accuracy. Precision represents the random system errors and quantifies how well a measurement can be carried out without a real reference value [33,34]. In the present paper, precision was estimated by the standard deviation (*SD*) between the data obtained by the load cell (y_i) and the average between the same ($\bar{y}$):

$$SD = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(y_i - \bar{y})^2} \quad (1)$$

Accuracy represents systematic errors and may be defined as the compliance degree of an average quantity related to its actual value [33,34]. Accuracy was estimated through the mean absolute deviation (*MAD*) between data obtained by the sensor (y_i) and the reference values (y_f), according to Eq. (2). In this case, the tare value before the vacuum and temperature control activation was used as a reference.

$$MAD = \frac{1}{N}\sum_{i=1}^{N}(y_i - y_f) \quad (2)$$

Experimental procedure for the dehydration process

Sample preparation

Avocados (*Persea americana*) from a local market in São Carlos (São Paulo State, Brazil) were used. The selection and cleaning process of the fruit was carried out as described by Dal-Bó and Freire [8]. The pulp was cut with a manual slicer, producing pieces with sizes of 25 x 25 x 15 mm (width x length x height). The samples were frozen on a PVC film on a tray in the lyophilizer condenser for 24 h at -30 °C. Each test employed a new fruit from the same batch that had been stored in an air-conditioned room for a maximum of three days. All the tests were performed in duplicate.

Moisture content

The moisture content of the fruit pulp was determined before and after the freeze-drying process, according to AOAC method 934.06 [35], by drying in a vacuum oven (Model TE-395, Tecnal®). The procedure was performed (in triplicate) for 24 h, at 70 °C, under reduced pressure of approximately 8 kPa.

The mass of each sample was measured using an analytical balance with a precision of 0.0001 g (A&D®, FR-200 MKII) before and after treatment in the vacuum oven. The moisture contents, based on wet and dry mass, were calculated using Eqs. (3) and (4), respectively:

$$X_{wb} = \frac{m_w}{m_o} \quad (3)$$

$$X_{db} = \frac{m_w}{m_f} \quad (4)$$

where X_{wb} is the moisture content on a wet basis [$kg_{water}/kg_{wet\ material}$]; X_{db} is the moisture content on a dry basis [$kg_{water}/kg_{dry\ solid}$]; m_0 and m_f are the masses [kg] of the sample before and after drying for 24 h in the oven, respectively; and m_w is the mass of water [kg] in the sample, determined as the difference between the initial and final masses of the sample.

Dehydration of the avocado pulp by freeze-drying

A slice of frozen avocado pulp was transferred using a clamp to the central region of the tray connected to the load cell, the system was closed, and the vacuum pump was activated. After starting the pump, the mean absolute operating pressure was 0.22±0.01 kPa, and the condensing surface temperature was -50.15±0.54 °C. It is important to highlight that these conditions are lower than those of triple point of water at 0.01°C and 0.611 kPa [36], which is generally required to perform the sublimation process [37,38]

The effect of heating temperature on the load cell measurements and the drying kinetics was investigated. For this, freeze-drying of the avocado pulp was performed using the sample tray with a heating temperature of 40 °C and without temperature control. The change in moisture content was monitored by recording the sample mass value every 3 s until constant mass. The reliability of the mass measurement system was evaluated under the conditions mentioned above by comparing the mass of the sample obtained by the developed system with that provided by an analytical balance (A&D®, Model FR-200 MKII) before and after the freezing-drying process.

Energy Aspect

To determine the effectiveness of the lyophilization system, with and without temperature

control, the values of specific moisture extraction rate (*SMER*), specific energy consumption (*SEC*), and moisture extraction rate (*MER*) were calculated by Eqs. (5), (6), and (7), respectively [39,40]:

$$SMER = \frac{m_w}{E_t} \quad (5)$$

$$SEC = \frac{1}{SMER} \quad (6)$$

$$MER = \frac{m_w}{t_d} \quad (7)$$

where m_w is the amount of water removed during drying, [kg water], E_t is the total energy supplied in the drying process, and [kJ]; e t_d is the drying time, [h].

The amount of energy required by the lyophilization process (primary and secondary drying stages) was calculated according to Huang *et al.* [41] and Dincer and Rosen [42] and includes six parts: the energy required for sublimation and water desorption, the energy required to raise the temperature of the material, condense the steam, evacuate the system, and provide heat by conduction (through the resistor) to heat the tray.

The values used for the energy calculations were based on the data provided by the equipment manual and on experimental data obtained for each studied condition. Enthalpy was determined from the thermodynamic tables [43]. It is important to highlight that although energy parameters analyzed for such a small system are hardly representative of an industrial process, they are useful o compare the effect of operational conditions on the energy saving of the freeze-drying process.

Data treatment and analysis

The moisture content (dry basis) and temperature data were used to obtain drying and temperature curves, as a function of freeze-drying time. The data were acquired every 3 s, but the graphs were plotted using intervals of 240 s to facilitate data visualization.

The dimensionless moisture contents were calculated as follows:

$$MR = \frac{X_t - X_{eq}}{X_0 - X_{eq}} \quad (8)$$

where X_t is the moisture content (dry basis) at a given instant [$kg_{water}/kg_{dry\ solid}$], and X_0 and X_{eq} are the moisture contents (dry basis) at the start of the process and dynamic equilibrium, respectively [$kg_{water}/kg_{dry\ solid}$].

Comparing the values of X_t or X_0 with the equilibrium moisture content, X_{eq} was relatively small (approximately zero), so the dimensionless moisture content could be simplified to Eq.(9), as described by Midilli *et al.* [44]:

$$MR = \frac{X_t}{X_0} \quad (9)$$

RESULTS AND DISCUSSION

Evaluation of the online mass measurement system

Figure 2 shows the mass data as a function of time for the blank tests under both analyzed conditions. When the system was evacuated, the drying chamber pressure reached equilibrium about 1 min after activating the pump, as displayed in Figure 3. The inset graph of Figure 2 shows that in both conditions studied, the mass recorded increased to 0.2 g after vacuum equilibrium was attained, followed by a slight decrease. This change was caused by the airflow during the evacuation, as described by Tribuzi and Laurindo [10]. Nevertheless, the mass measurement increased again after this event and reached a plateau-like state. In the test without tray heating temperature control, the mass measurement increased and stabilized in values close to 0.2 g, while for the test with heating temperature control, the stabilization was observed in values similar to 0.35 g. The difference between the curves obtained with and without temperature control can be attributed to the influence of temperature on the load cell. Temperature affects the load cell output by altering the sensitivity of the load cell [45]. Note that in the process without tray heating temperature control, the masses obtained for the duplicates presented greater oscillations because the ambient temperature influenced them due to radiation heat transfer (even though the tests were performed in an air-conditioned room, closed to the sunlight, under artificial light source).

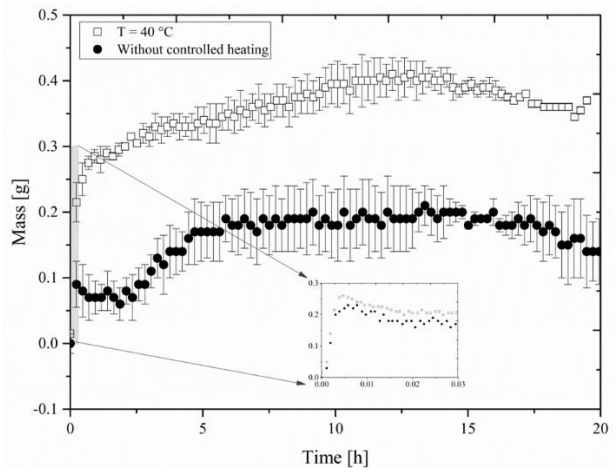


Figure 2. Mass, as a function of time, is in the blank test to evaluate the data acquisition system.

Using the data shown in Figure 2, the overall precision and accuracy of the mass measurement system could be determined as 0.107 g and 0.240 g, respectively. Considering that 8.8 g of fruit was used, on average, and the weight variation due to the drying process was approximately 7.4 g, the error introduced by the system's accuracy represents only 3.2% of the mass variation. Thus, the system developed can be considered precise and accurate for determining the sample mass variation induced by drying. Nevertheless, Figure 2 shows that the data obtained were not randomly distributed around zero, and the deviations observed were reproducible (low confidence intervals). Thus, blank curves were obtained (in duplicate) for the different operating conditions before and after each sample dehydration test to increase the system's accuracy and correct the avocado pulp mass values obtained during the freeze-drying process. The results of using blank curves to correct the mass of the material will be shown in the next section.

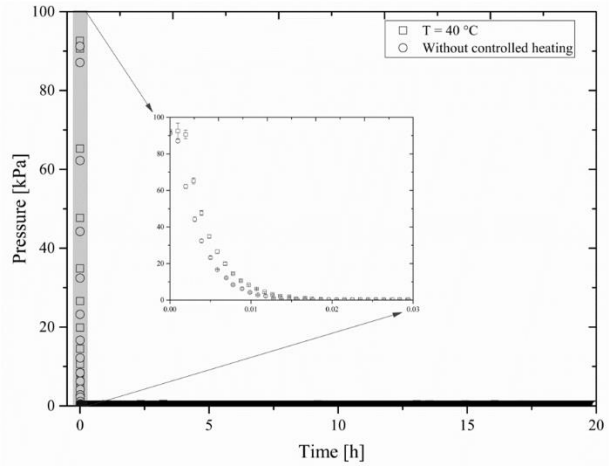


Figure 3. Pressure in the freeze-drying system as a function of drying time.

The deviations from the reference value (zero) and oscillations observed can be attributed to many factors. Typical load cells use strain gauges that deform when a load is applied to the contact area, consequently increasing the resistance [46–48]. As Hernandez [49] and Muller *et al.* [50] pointed out, temperature, linearity, hysteresis, repeatability, and/or flux density influence the signal obtained using a strain gauge. Noise from the electricity supply network can also affect the ability of a load cell to produce the same value when the same weight is applied [49,51]. It is because the response given by the amplifier module is a voltage difference, depending on the amplifier gain. In addition to these factors, vibrations caused by the vacuum pump and oscillations in the degree of vacuum affect the precision of the mass measurement.

Freeze-drying of the avocado pulp

After the blank assays, the avocado pulp was dried with either an unheated tray or heated at 40 °C. The air temperature, pressure, and relative humidity of the environment outside of the chamber were monitored during the freeze-drying tests, with mean values of 23.71 ± 0.46 °C, 91.99 ± 0.10 kPa, and 37.18 ± 2.35%, respectively. Figure 4 shows the results of the avocado pulp mass as a function of drying time.

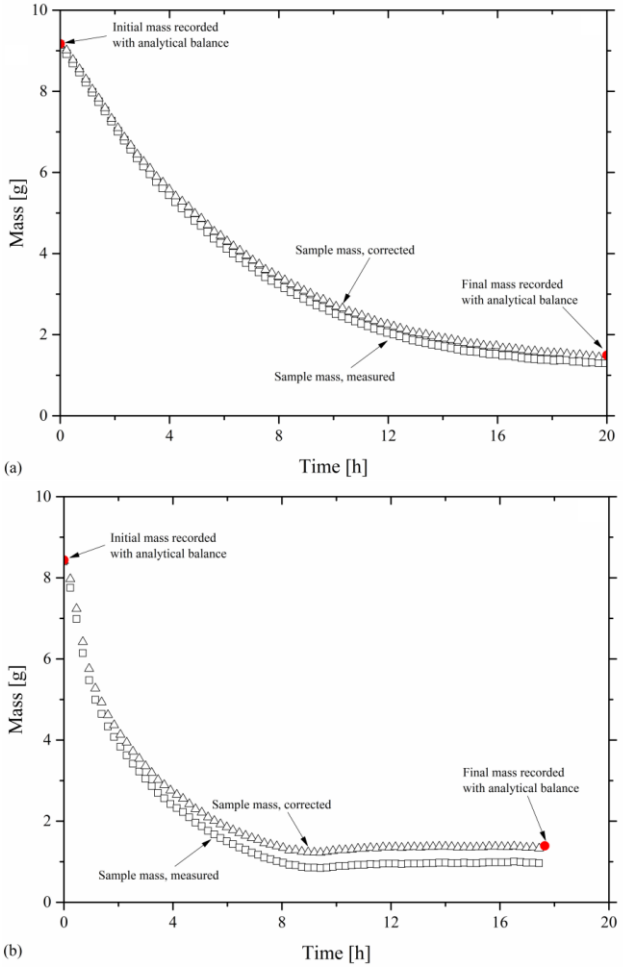


Figure 4. Sample mass as a function of drying time. Drying conditions: (a) without temperature control of the heating tray; and (b) heating plate temperature at 40 °C.

As shown in Figure 4b, the heating temperature greatly influenced the mass obtained with the load cell and registered with the microcontroller in Arduino®. Note that the difference between curves is greater in the final drying period, which is justified due to the lower mass of the sample. Greater masses added may reduce errors during data collection [10].

The avocado pulp's mass value was measured by an analytical balance (accuracy of ± 0.0001 g) before and after the freezing-drying process to check the system's reliability. The values obtained by the analytical balance were marked in Figure 4 with red dots. A Student's t-test was performed between the values measured by the analytical balance and those recorded by the data acquisition system, with and

without the application of correction curves, before and after the freezing-drying process, for both temperature control conditions tested. The results are presented in Table 1 and showed, with 95% of reliability, that there is no statistically significant difference (*p-value* > 0.05) between the data obtained by the analytical balance and by the acquisition system before the freezing-drying process, for both temperature conditions, with or without curves of corrections. Nevertheless, after the freezing-drying process, the results showed that values obtained without the blank curve correction were statistically significantly different from those obtained by the analytical balance for both temperature control conditions. On the other hand, the application of correction curves turned the values obtained by the data acquisition system statistically equal to those obtained by the analytical balance. These results reinforce the need for corrections by blank curves to obtain reliable values of the mass of material during the freezing-drying process.

Table 1. Results of probability value (p-value) of the Student's t- test.

Temperature control	Blank curve correction	Freezing drying process	
		Before	After
No	Yes	0.388	0.089
	No	0.921	0.023
Yes (40 °C)	Yes	0.227	0.134
	No	0.360	0.006

Figure 5 shows the temperature of the sample, with tray heating temperature at 40 °C and without temperature control, as a function of time, together with the corresponding standard deviations. During the freeze-drying process, the mean experimental errors for the temperature curves of the sample and the heating plate were ±1.40 and ±0.21 °C (Figure 5a) and ±2.28 and ± 0.52 °C (Figure 5b), respectively. Greater deviations were observed for the sample temperature due to the variability of the composition of the same fruit. As shown in Figure 5a, the sample temperature remained low (during the sublimation stage), despite the heat supply, due to the high heat of sublimation of the ice. The shift to the desorption step occurred when the ice was sublimated entirely and the sample temperature approached the heating temperature. The start of the transition from sublimation to desorption was observed when the mean sample temperature reached 21 °C (after 8 h). When the heat was not supplied during the freeze-drying process, the temperature of the sample showed a constant increase (Figure 5b). The shift from sublimation to desorption occurred at the end of the process (after around 18 h of freeze-drying), reaching a mean equilibrium temperature of 21 °C. This result shows that the freeze-drying chamber was not completely thermally insulated from the external environment, which hampers the possibility of transferring the drying kinetics results obtained, and constitutes a point of improvement in the system. Nevertheless, as shown below, the proposed methodology allowed for obtaining the materials' highly reproducible drying kinetics curves, meeting the final moisture content control goal.

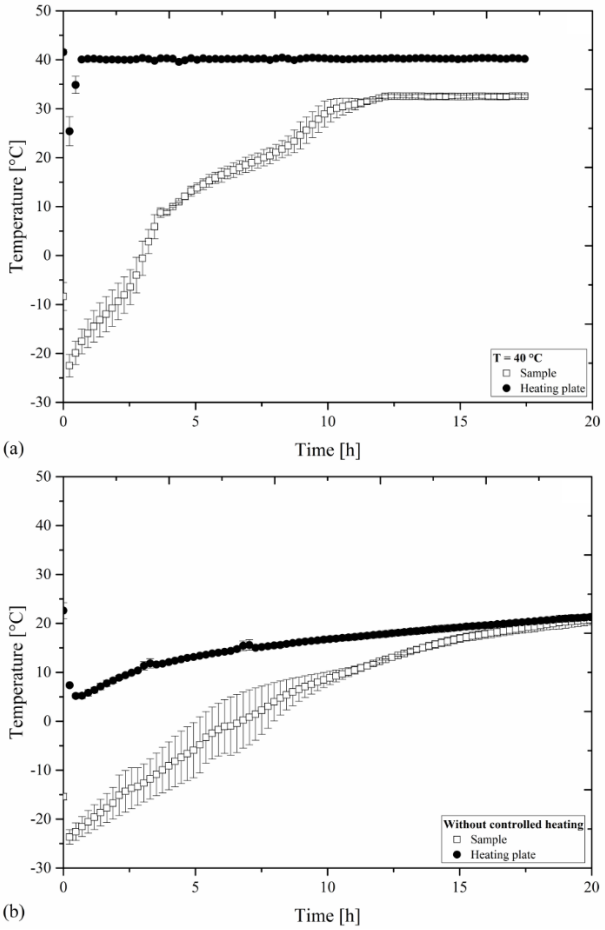


Figure 5. The temperature of the sample, as a function of time, during the freeze-drying process: (a) with heating at 40 °C and (b) without heating temperature control.

The controlled supply of heat during the freeze-drying process is essential for decreasing the drying time, consequently reducing the energy expenditure associated with the application of a vacuum. However, a substantial increase in the plate temperature is not recommended because the region of the sample in direct contact with the plate could overheat, compromising the product's quality and altering its visual appearance. Furthermore, as pointed out by Levi and Karel [52] and Pikal and Shah [53], exceeding certain temperature limits could cause the sample to collapse, with the sealing of capillaries, reduction of the pore size, loss of structure, decreased dehydration, and swelling.

The mean moisture content of *natural* avocado

pulp was 0.86 ± 0.01 $kg_{water}/kg_{wet\ material}$ (6.12 ± 0.51 $kg_{water}/kg_{dry\ solid}$, on a dry basis). Figure 6 shows the results for the dimensionless moisture content (dry basis) as a function of the avocado pulp drying time. The curves presented satisfactory reproducibility, with a mean standard deviation of ± 0.03, which allows for controlling the final moisture content of the freeze-dried sample produced. The application of the proposed system to produce freeze-dried samples can be observed in more detail elsewhere [8]. The authors pointed out good reproducibility and efficiency in temperature control, allowing to study the effect of drying temperature on the drying time and quality of the final product.

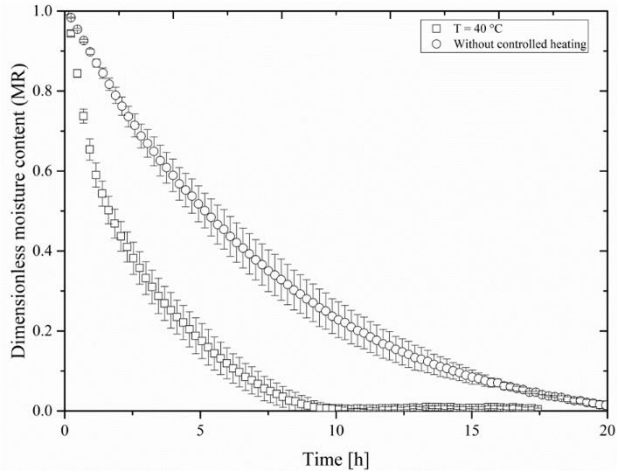


Figure 6. Dimensionless moisture content, as a function of the avocado pulp freeze-drying time, with and without tray heating temperature control.

As expected, the controlled heat supply during the freeze-drying accelerated the dehydration process, decreasing the drying time by about 0.63 times. The freeze-drying process without heating control reached dynamic equilibrium after around 20 h. In previous work, Souza *et al.* [54] found that equilibrium was reached after around 13 h. This difference could have been due to factors related to the freezing of the sample, the operating conditions, the freeze-drying system, and the qualities of the raw materials used.

As shown in Figure 7, higher drying rates were observed when the heat was supplied in a controlled manner during the process. The driving force in freeze-drying is the temperature difference and, therefore, the vapor pressure difference between the sublimation surface of the sample and the ice layer on the condenser. Thus, a greater temperature difference is expected to provide higher drying rates. Besides, with the addition of excess heat, there was the initiation of the induction period, with the temperature of the product increasing, consequently increasing the drying rate. The average duration of these periods was around 21 min and 7 min for the processes with and without the tray heating temperature control, respectively.

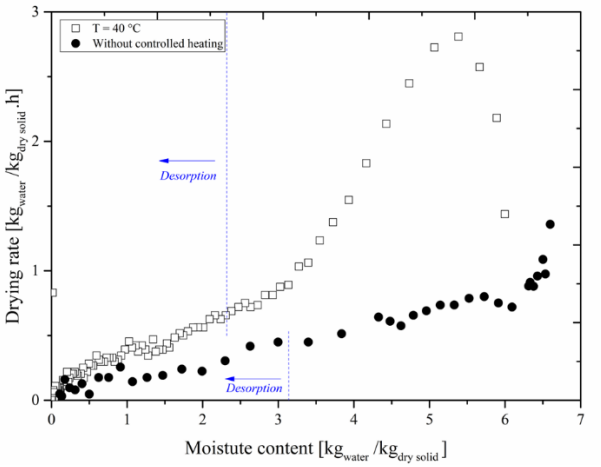


Figure 7. Drying rates as a function of moisture content (dry basis).

After the induction period, there was a period during which the drying rate decreased, which is characteristic of biological materials. During this period, the average drying rates were 1.42 and 0.56 $(kg_{water}/kg_{dry\ solid}).h^{-1}$ for the processes with and without tray heating temperature control, respectively. The material presented greater water loss during the sublimation stage since the water was not bound to the molecular structure of the pulp [55], so the drying rate was faster. Only residual water was removed during the desorption stage, so the drying rate was slower.

Energy Aspect

Table 2 shows the freeze-drying process's drying time and energy indexes, with and without heating temperature control. The specific moisture extraction rate indicates the energy efficiency in drying. The results showed that the process with temperature control had a higher *SMER* index, indicating a greater amount of water removed and, consequently, better thermal efficiency than without temperature control.

A higher value of specific energy consumption for the process without temperature control was also reported by Liu *et al.* [57], and this may be related to the long operational time linked to the low pressure of the drying chamber and the low-temperature state of the cold trap, which result in high energy consumption per kilogram of water removed [1,57,58].

By analyzing the valid energy consumption of the system, it is observed that despite the controlled heat supply, the process with temperature control showed less energy required to operate. It is due to the shorter operational time and the balance between the sublimation and desorption steps. In general, as shown in Table 2, the lyophilization process covered in this study showed better energy indexes when compared to

Table 2. Energy indexes of freeze-drying avocado pulp.

	T = 40 °C	Without temperature control	Black chokeberry puree[1]	Purple basil leaves[2]
Drying time [h]	12.13	19.48	3.00	8.00
Energy consumption [kWh]	0.0398	0.0492	1.36	5.67
SMER [kg/kWh]	0.1943	0.1601	0.016	0.001
SEC [kWh/kg]	5.15	6.24	61.72	709.09
MER [kg/h]	6.37×10^{-4}	4.04×10^{-4}	72.5×10^{-4}	10×10^{-4}

[1][39]; [2][56].

CONCLUSION

This paper showed in extensive detail the adaptation methodology of a laboratory-scale freeze dryer to monitor the production process of freeze-dried samples. The following conclusions can be drawn from this study:

The methodology described permitted the development of a precise and accurate data acquisition system for measuring the temporal changes in the sample mass and temperature and chamber pressure variations, estimating the drying rates, temperature heating behavior, and energy aspects of the production process of freeze-dried samples.

The use of higher temperatures (40 °C) in the freeze-drying process increased the errors involved in measuring the mass of material; nevertheless, the application of correction blank curves statistically significantly reduced those errors.

The controlled heat supply accelerated the freeze-drying process and enabled the evaluation of the effect of temperature on the drying rate. Besides, the temperature control decreased the specific energy consumption of the process.

Overall, the methodology described in this paper can be extended to adapting similar freeze-drying equipment, using low-cost devices, making the equipment more efficient and automated, especially for producing freeze-dried samples for research purposes.

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NAUČNI RAD

RAZVOJ SISTEMA ZA PRAĆENJE PROCESA PROIZVODNJE UZORAKA OSUŠENIH ZAMRZAVANJEM: JEDNOSTAVAN I JEFTIN PRISTUP

Prikupljanje podataka iz procesa sušenja zamrzavanjem je važno za dobijanje liofiliziranih uzoraka sa željenim konačnim sadržajem vlage u različitim radnim uslovima. Ovo istraživanje opširno predstavlja jednostavnu i jeftinu metodologiju za implementaciju sistema za prikupljanje podataka u laboratorijskoj sušari zamrzavanjem. Rezultati su pokazali da više temperature sušenja (40 °C) povećavaju greške u merenju mase materijala; ipak, primena ispravnih praznih krivih je statistički značajno smanjila te greške. Generalno, razvijeni sistem je obezbedio precizna i tačna merenja vremenskih promena mase i temperature uzoraka, kao i varijacije pritiska u komori, omogućavajući praćenje procesa proizvodnje zamrzavanjem osušenih uzoraka sa niskim sadržajem finalne vlage..

Ključne reči: liofilizacija, sušenje, zamrzavanje, temperatura grejanja, Arduino, avokado.