

## PAPER

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## Potential of vacuum impregnation and osmotic dehydration techniques in producing jaggery-fortified apple snacks

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Fruits are nutrient-rich, highly perishable goods which contribute to postharvest losses and waste. The food industry continues the search for processing methods that allow for the manufacturing of attractive and convenient fortified fruits while extending their shelf life. To meet the present consumer demands for more nutritious and sustainable food products, innovative or revisited food processing techniques need to be explored. In the present work, jaggery is proposed as a non-conventional osmotic agent to produce fortified apple snacks through the combination of vacuum impregnation (VI) and osmotic dehydration (OD) techniques and further stabilization via convective hot air-drying (HAD) or freeze drying (FD). Physicochemical and antioxidant attributes of intermediate and final products were analyzed to evaluate the potential of these techniques to introduce jaggery bioactive constituents in the apple matrix. The results confirmed that the antioxidant properties of jaggery may be incorporated into the tissue by both VI and OD, especially with progressive OD (pOD) in solutions from 30 to 50 Brix degrees. Stabilization through HAD at 60 °C significantly enhanced the antioxidant properties of jaggery-enriched snacks (total phenols:  $11.0 \pm 0.6$  (pOD HAD) and  $8.0 \pm 0.6$  (VI HAD) vs.  $6.3 \pm 0.12$  (HAD) mg GAE per g dry product), whereas FD maintained natural and incorporated antioxidants (total phenols:  $10.8 \pm 0.4$  (pOD FD) and  $6.2 \pm 0.9$  (VI FD) vs.  $6.5 \pm 0.2$  (FD) mg GAE per g dry product). Optical and textural properties were affected by the addition of jaggery and processing techniques. Replacing intercellular air with liquid reduced luminosity, which increased after dehydration, especially through FD. In conclusion, jaggery or non-centrifugal cane sugar is proposed as a healthier osmotic agent to produce more nutritious and sustainable apple snacks by applying matrix engineering techniques such as vacuum impregnation and osmotic dehydration, followed by hot air-drying or freeze-drying stabilization.

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## Sustainability spotlight

To reduce postharvest losses and waste, the food industry needs to develop sustainable processing methods that allow for the extension of fruit shelf life while manufacturing attractive and convenient nutrient-rich products. A combination of matrix engineering and drying techniques is proposed to produce jaggery-fortified apple snacks. This study addresses Sustainable Development Goal 2 (SDG2: end hunger, achieve food security and improved nutrition and promote sustainable agriculture) by producing fortified apple snacks from perishable fruit, thus reducing food waste, and promoting access to nutrient-dense convenient food. In addition to the mitigation of fruit waste through fruit snack manufacturing, the use of vacuum impregnation and osmotic dehydration techniques reduces the energy requirements of drying processes, thus contributing to more sustainable processes addressing Sustainable Development Goal 12 (SDG12: ensure sustainable consumption and production patterns).

## Introduction

Fruits contain nutrients and bioactive compounds, making them an important part of the human diet. However, they are highly perishable goods and lead to postharvest losses and waste, thus resulting in serious environmental problems<sup>1</sup> and nutritional losses.<sup>2</sup> Both fresh and processed fruits are considered good preferences from a nutritional point of view.

Therefore, the food industry continues the search for processing methods that allow for the manufacturing of attractive and convenient fortified fruits while extending their shelf life.<sup>3</sup> In this context, innovative or revisited food processing techniques have gained importance to offer alternatives to meet present consumer demands for more sustainable and healthy diets, as proposed by the FAO.<sup>4</sup>

Osmotic dehydration (OD) and vacuum impregnation (VI) are used to improve the nutritional, sensory and functional values of food products, particularly when used as a pretreatment in preserving operations, such as drying or freezing.<sup>5</sup>

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Among dehydration methods, the combination of osmosis and drying has proven to be an efficient strategy for improving the quality of fruits and vegetables.<sup>6</sup> In addition, OD and VI might be used to intentionally introduce bioactive compounds that are not naturally present in the product into the food matrix. Although OD involves a two-way mass transfer process that couples dehydration with an inflow of solutes and other desired nutritional substances into the plant material,<sup>3</sup> VI is a process of fluid replacement in porous materials *via* vacuum pressure and subsequent atmospheric pressure reestablishment, which allows for the direct introduction of solutes and other desired compounds into the porous structure of foodstuffs, thereby enhancing mass transfer rates through hydrodynamic transport.<sup>5,7</sup> The ability to control these processes and introduce desired nutritional substances into plant material presents remarkable opportunities for the development of sustainable functional foods.<sup>7,8</sup> From a sustainable perspective, OD and VI are non-thermal treatments and are considered low-energy demanding operations. In addition, combined with further drying or freezing, they are known to reduce energy consumption.<sup>8,9</sup>

A combination of matrix engineering technologies, such as OD and VI, with drying operations is proposed to produce appealing and nutritious functional fruit snacks. This combination not only enhances the flavor and texture of the product but also retains essential nutrients, transforming fruits into convenient on-the-go foods without compromising or ideally increasing their nutritional value. For example, it has been used to produce calcium-rich apple and pineapple snacks and juice-enriched apple snacks, or to introduce and stabilize probiotic microorganisms in the food matrix.<sup>10–13</sup> However, jaggery or non-centrifugal sugar is an unconventional natural sweetener that used to be the most common form of sugarcane product before the advent of refined sugar.<sup>9</sup> It is still commonly consumed across the tropics and subtropics, where it is produced and used as part of traditional medicine.<sup>14</sup> In contrast to refined sugar, jaggery contains several bioactive compounds that make it interesting for functional food development. Apart from being richer in vitamins and minerals, non-refined sugarcane products are particularly rich in phenolic constituents, especially flavonoids, and exhibit antioxidant and antimicrobial effects.<sup>14–17</sup> As reported in various studies, various health promoting effects are attributed to sugarcane extracts, including antiproliferative properties against cancer cell lines, stimulation and regulation of the immune system, protection against hepatic damage, recovery of intestinal function, anti-thrombotic and anti-stress properties, protective role against DNA damage, growth stimulation, or prevention from hypertension and diabetes disorders.

Jaggery consists of sugarcane juice concentrated by evaporation. Other fruit or vegetable juices have been proposed as vacuum impregnation solutions to produce functional foods.<sup>18–20</sup> In contrast to other fruit juices, jaggery contains sugar mostly in the form of sucrose, and it is presented as a dehydrated powdered product in the case of granulated jaggery.<sup>14</sup> This makes granulated jaggery an interesting alternative for the formulation of osmotic solutions. Osmotic

solutions to be used in OD and VI processes could benefit from jaggery's healthier properties. However, the use of jaggery as an osmotic agent in VI and OD operations has rarely been exploited in the literature.<sup>21</sup> In the present work, granulated jaggery is proposed as an osmotic agent to produce fortified apple snacks by VI and OD with further stabilization by convective hot air-drying (HAD) or freeze-drying (FD). The physicochemical and antioxidant attributes of intermediate and final products are analyzed to evaluate the potential of these techniques to produce more nutritious and sustainable apple snacks.

## Materials and methods

### Raw materials

Apples (*Malus domestica* cv. Granny Smith) used in the present study were purchased from a local market in Valencia, Spain. This variety was chosen owing to its high homogeneity and porosity, which present excellent attributes for VI. The apples were washed with tap water and cut into 5 mm thick slices, which were then transformed into rings with internal and external diameters of 20 mm and 65 mm, respectively. Granulated jaggery and white sugar used to prepare the osmotic solutions were obtained from a local market.

### Snack manufacturing techniques

VI and OD were used alone or combined to obtain intermediate products that were further stabilized by HAD or FD. Details of the conditions of the operations used and the properties analyzed in each case are presented in the subsequent sections. Intermediate VI and OD products were compared to the fresh apple, whereas the controls for dehydrated snacks consisted of HAD and FD samples with no pretreatment.

### Vacuum impregnation (VI)

VI was carried out by soaking the apple rings in isotonic impregnation solutions (1 : 4 mass ratio) and introducing them into a vacuum chamber (Heraeus Vacuum Oven, Thermo Fisher Scientific Inc.) connected to a vacuum pump (Gardner Denver Thomas GmbH Welch Vacuum, Fürstentfeldbruck, Germany), where a pressure of 50 mbar was applied for 10 min. After that, atmospheric pressure was restored, and the samples were soaked in the solutions for 10 more min.

Impregnation solutions consisted of isotonic solutions (~250 g sugar per L) prepared with combinations of white sugar and jaggery using the following jaggery percentages with respect

**Table 1** Composition of impregnation solutions isotonic with the apple tissue, prepared with different percentages of granulated jaggery (0–100% GJ) as a replacement to white sugar

	White sugar (g)	Jaggery (g)
0% GJ	250	0
25% GJ	187.5	62.5
50% GJ	125	125
100% GJ	0	250



to total sugar added: 0, 25, 50 and 100% (Table 1). The impregnation properties of the solutions in the apple tissue were evaluated using VI parameters, the procedure, equipment and equations defined by Salvatori *et al.* (1998).<sup>22</sup> Hence, the following parameters were obtained:  $X_1$ , volumetric fraction occupied by the impregnation liquid after the vacuum step ( $\text{m}^3$  solution per  $\text{m}^3$  fresh tissue);  $X$ , volumetric fraction occupied by the impregnation liquid after the atmospheric pressure step ( $\text{m}^3$  solution per  $\text{m}^3$  fresh tissue);  $\gamma_1$ : relative volumetric deformation after the vacuum step; and  $\gamma$ : relative volumetric deformation after the atmospheric pressure step. Experiments were performed at room temperature.

### Osmotic dehydration (OD)

OD was used as an alternative or complementary technique to incorporate the bioactive compounds of interest contained in jaggery into the apple structure. For OD experiments, 100% jaggery solutions were used in all cases, and different dehydration strategies were examined: one step OD in a 50 Brix degrees jaggery solution during 3 h (OD), or progressive OD in a 30, 40 and 50 Brix degrees jaggery solution (pOD), maintaining the samples in each solution for 1 h. Experiments were conducted at room temperature.

### Convective drying (HAD) and freeze drying (FD)

Convective drying (hot air drying, HAD) was carried out in a CLW 750 TOP+ pilot plant tray dryer (Pol-Eko-Aparatura SPJ, Vladislava, Poland) with cross flow air at  $2 \text{ m s}^{-1}$  and  $60^\circ\text{C}$  for 24 h to reduce water activity to values that guarantee snack stability.<sup>23</sup> Freeze-drying (FD) was performed by freezing samples at  $-40^\circ\text{C}$  for 24 h in a CVN-40/105 Matek freezer and further sublimating at  $-45^\circ\text{C}$  (condenser temperature) and 0.1 mbar for 24 h in a 6–80 Telstar Lioalfa pilot plant scale freeze-dryer.

### Analytical determinations

**Apparent density and viscosity.** Impregnation solutions were evaluated in terms of apparent density and viscosity. Apparent density ( $\rho$ ,  $\text{g cm}^{-3}$ ) was obtained using a pycnometer with distilled water used as a reference at  $25^\circ\text{C}$ . Viscosity ( $\mu$ , Pa s) was determined using a rotatory rheometer (HAAKE, Rheo Stress RS1, Germany) at  $20^\circ\text{C}$  and a speed gradient between 0 and  $70 \text{ s}^{-1}$  (20 cycles of 10 s). Viscosity values were obtained by applying Newton's law ( $\tau = \mu\gamma$ ) because all solutions showed a Newtonian behaviour.

**Moisture content ( $x_w$ ), water activity ( $a_w$ ) and soluble solid content ( $x_{ss}$ ).** Moisture contents of fresh apples and derived products were measured by vacuum drying following the AOAC official method (method 20.013, AOAC, 1990),<sup>24</sup> whereas water activity was measured at  $25^\circ\text{C}$  in a calibrated dew point hygrometer (Decagon Aqualab model CX-2, Pullman, WA, USA). Brix degree measurements were obtained with a benchtop thermostatic refractometer (ABBE ATAGO 3-T, Japan) at  $20^\circ\text{C}$ .

**Antioxidant properties.** Antioxidant properties were assessed by determining total phenols, total flavonoids, and DPPH and ABTS radical scavenging activities. Determinations were

performed on extracts obtained from a known amount of sample (2 g for non-dried apple samples and 0.35 g for dried ones) and 20 mL of an 80 : 20 methanol : water solution used as the solvent. Extraction was conducted by stirring the mixture for 1 h and subsequently centrifuging it at 10 000 rpm for 5 min in an Eppendorf centrifuge 5804/5804R (Eppendorf SE, Hamburg, Germany) to obtain a clear supernatant, which was used for further measurements.

The total phenolic content was measured using the spectrophotometric method of the Folin–Ciocalteu reagent.<sup>25</sup> An aliquot of 0.125 mL of the extract was mixed with 0.5 mL of distilled water and 0.125 of the Folin–Ciocalteu reagent (Sigma Aldrich). The mixture was allowed to react for 7 min in the dark before adding 1.25 mL of a 7% sodium carbonate solution to stop the reaction and 1 mL of distilled water. Absorbance was measured at 760 nm using a spectrophotometer (Helios Zeta UV/vis, Thermo Scientific, UK) after 90 min in the dark. The results were presented in mg of Gallic Acid Equivalents (GAE) per g of fresh or dried sample. The total flavonoid content was obtained using the colorimetric aluminum chloride method described by Luximon-Ramma (2002),<sup>26</sup> which consisted of vigorously mixing 1.5 mL of the extract with 1.5 mL of aluminum chloride solution (2% w/v in methanol) and measuring the absorbance at 368 nm using apigenin as the standard (purity  $\geq 95\%$ , Sigma-Aldrich). Results were given in mg of apigenin equivalents (AE) per gram of fresh or dried sample.

**Antiradical DPPH (1,1-diphenyl-2-picryl hydrazyl, DPPH-) activity assay** was based on the method developed by Brand-Williams *et al.* (1995)<sup>27</sup> and consisted of adding 0.05 mL of the extract to 2.95 mL of a 0.06 mM DPPH-methanol solution. The ability to scavenge the ABTS<sup>+</sup> cation (2,2'-azobis-3-ethyl benzothiazoline-6-sulphonic acid) was measured, as described by Re *et al.* (1999).<sup>28</sup> The radical ABTS<sup>+</sup> was released by reacting 7 mM of ABTS with potassium persulfate (2.45 mM) for 16 h at room temperature and in the dark. ABTS<sup>+</sup> was mixed with phosphate buffer (pH 7.4) to reach an absorbance of  $0.70 \pm 0.02$  at 734 nm. An aliquot of 100  $\mu\text{L}$  of the sample was added to 2900  $\mu\text{L}$  of the solution ABTS<sup>+</sup> in phosphate buffer, and the absorbance of the samples was read after 7 min. Antiradical activities were both expressed in mg of Trolox Equivalent (TE) per gram of fresh or dried sample.

**Optical properties.** The colour of impregnation and osmotic solutions and apple products was measured using a Minolta CM 3600D spectrophotometer (Konica Minolta Sensing, Inc., Japan). Colour coordinates of the CIE  $L^*a^*b^*$  colour space were obtained by reflectance from the absorption spectrum provided by the equipment in the 380–770 nm range with D65 illuminant and  $10^\circ$  observer. The readings were made on black and white backgrounds by applying the Kubelka–Munk theory for multiple scattering of reflection spectra.<sup>29,30</sup> Liquid samples were placed in 25 mL standardized plastic cuvettes of 2 cm optical path length, whereas solid samples were directly placed on the sample holder. Colour attributes of chrome ( $C_{ab}^*$ ) and hue angle ( $h_{ab}^*$ ) were obtained from the CIE  $L^*a^*b^*$  colour coordinates by applying the following equations  $C_{ab}^* = (a^{*2} + b^{*2})^{1/2}$  and  $h_{ab}^* = \arctg(b^*/a^*)$ , respectively.



Colour differences were calculated using the following equation:  $\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ .

**Mechanical properties.** The mechanical behavior of fresh apples and snacks was measured using a puncture test on a Universal Texture Analyser (Stable MicroSystems, TA.XT2 Plus, Ltd, Godalming, UK) and the software Texture Exponent 32 v. 6.1.2.0. A cylindrical 2 mm diameter punch was used for the analyses, with the following assay conditions: rate 2.0 mm s<sup>-1</sup>, activation force 0.049 N, distance run 10 mm, and charge cell 50 kg. The parameters obtained from the puncture test were the maximum force ( $F_{\max}$ , N) and the distance at which the maximum force was reached ( $d_{\max}$ , mm). Deformation at the maximum force point was obtained from  $d_{\max}$  considering the sample thickness ( $d_{\max}/l$ ).

**Statistical significance of results.** Results were statistically analyzed using Statgraphics Centurion software (Centurion XVII.I version, StatPoint Technologies, Inc., Warrenton, VA, USA) with a confidence level of 95% ( $p$ -value = 0.05). The data were processed by performing simple and multifactor ANOVA. All analytical determinations were performed at least in triplicate.





## Results

### Properties of impregnation solutions

The characterization of the impregnation solutions is summarized in Table 2. As observed, replacing white sugar with jaggery did not significantly affect the general physicochemical attributes. Water activities were similar for the four solutions formulated, which were isotonic with the apples used in the study ( $a_w = 0.989$ ). This implies that the only mechanisms involved in mass transfer during VI are pressure gradients.

Other physicochemical parameters, such as Brix degrees, apparent density ( $\rho$ ), and viscosity ( $\mu$ ), were also similar among the four impregnation solutions prepared. All impregnation liquids showed a Newtonian behaviour and exhibited viscosity values slightly higher than pure water, indicating a certain ease of being impregnated in the cellular tissue. It has been reported that low viscosity values (smaller than 3–3.5 mPa s) favor liquid impregnation in contrast to tissue deformation and further relaxation.<sup>31</sup> However, impregnation parameters exhibited certain differences depending on the amount of jaggery participating in the solution. The main differences were found in parameter  $X$ , *i.e.* the volumetric fraction of the impregnation solution that is introduced in the apple matrix at the end of the atmospheric step. The results indicated poorer impregnation properties of solutions containing jaggery. This could be explained by the presence of other compounds different from sucrose, especially insoluble ones, which might accumulate on the surface of the sample and obstruct open pores, thus hindering liquid inflow. However, because both  $X$  and  $X_1$  presented positive values, the impregnation liquid entered during both the vacuum ( $X_1 > 0$ ) and the atmospheric ( $X > 0$ ) stages. As for volumetric deformation, the positive values during the vacuum step ( $\gamma_1 > 0$ ) indicate expansion, whereas the negative values at the end of the atmospheric one ( $\gamma_1 < 0$ ) suggest a certain contraction of the apple tissue. In any case, no significant differences among the samples were obtained for the deformation parameters. To the best of our knowledge, this is the first time that the VI properties of jaggery-formulated solutions have been reported. Nevertheless, the response of Granny Smith apples to VI has been previously studied for other impregnation solutions. The impregnation parameters of apple

**Table 2** Physicochemical, impregnation, and optical properties of impregnation solutions isotonic with the apple tissue prepared with different percentages of granulated jaggery (0–100% GJ) as a replacement to white sugar. Mean (standard deviation) of three replicates<sup>a</sup>

	0% GJ	25% GJ	50% GJ	100% GJ
$a_w$	0.986 (0.003) <sup>b</sup>	0.984 (0.003) <sup>a</sup>	0.985 (0.003) <sup>b</sup>	0.985 (0.003) <sup>ab</sup>
Brix	24.81 (0.12) <sup>b</sup>	25.83 (0.12) <sup>c</sup>	24.70 (0.10) <sup>b</sup>	24.47 (0.06) <sup>a</sup>
$\rho$ (g cm <sup>-3</sup> )	1.10157 (0.00004) <sup>c</sup>	1.1065 (0.00010) <sup>d</sup>	1.10103 (0.00002) <sup>b</sup>	1.1002 (0.0002) <sup>a</sup>
$\mu$ (mPa s)	2.57 (0.06) <sup>a</sup>	2.57 (0.06) <sup>a</sup>	2.57 (0.06) <sup>a</sup>	2.57 (0.06) <sup>a</sup>
$X$	0.26 (0.02) <sup>b</sup>	0.18 (0.02) <sup>a</sup>	0.192 (0.0014) <sup>a</sup>	0.159 (0.004) <sup>a</sup>
$X_1$	0.04 (0.011) <sup>a</sup>	0.033 (0.002) <sup>a</sup>	0.05 (0.02) <sup>a</sup>	0.04 (0.011) <sup>a</sup>
$\gamma$	-0.05 (0.07) <sup>a</sup>	-0.040 (0.006) <sup>a</sup>	-0.053 (0.0010) <sup>a</sup>	-0.07 (0.012) <sup>a</sup>
$\gamma_1$	0.01 (0.08) <sup>a</sup>	0.023 (0.007) <sup>a</sup>	0.007 (0.005) <sup>a</sup>	-0.02 (0.04) <sup>a</sup>
$L^*$	55.79 (0.05) <sup>c</sup>	28.8 (0.11) <sup>b</sup>	26.5 (0.2) <sup>a</sup>	26.38 (0.02) <sup>a</sup>
$a^*$	-0.61 (0.07) <sup>a</sup>	4.17 (0.04) <sup>d</sup>	1.94 (0.04) <sup>c</sup>	1.00 (0.02) <sup>b</sup>
$b^*$	0.41 (0.03) <sup>a</sup>	6.7 (0.2) <sup>d</sup>	2.92 (0.09) <sup>c</sup>	2.51 (0.05) <sup>b</sup>
$h_{ab}^*$	146 (5) <sup>c</sup>	58.0 (0.7) <sup>a</sup>	56.5 (0.7) <sup>a</sup>	68.4 (0.5) <sup>b</sup>
$C_{ab}^*$	0.73 (0.05) <sup>a</sup>	7.9 (0.2) <sup>d</sup>	3.51 (0.09) <sup>c</sup>	2.71 (0.05) <sup>b</sup>
$\Delta E$	—	28.13 (0.06) <sup>a</sup>	29.5 (0.15) <sup>b</sup>	29.53 (0.02) <sup>b</sup>
Visual appearance				

<sup>a</sup> a, b, c, d different letters in the same row indicate statistically significant differences at the 95% level ( $p$ -value  $\leq 0.05$ ).





cylinders (20 mm diameter  $\times$  20 mm height) with sucrose isotonic solutions have been reported as  $X_1 = -0.042 \pm 0.003$ ,  $\gamma_1 = 1.7 \pm 0.3$ ,  $X = 0.19 \pm 0.015$  and  $\gamma = -0.6 \pm 1.2$ ,<sup>32</sup> whereas the impregnation parameters for apple slices (5 mm thick) immersed in clementine juice enriched with *Lactobacillus salivarius* were  $X_1 = 0.150 \pm 0.010$ ,  $\gamma_1 = 0.091 \pm 0.003$ ,  $X = 0.19 \pm 0.02$  and  $\gamma = 0.012 \pm 0.007$ .<sup>33</sup>

Jaggery, compared to white sugar, is characterized by its brownish colour, which is provided by the natural constituents of sugarcane, mainly flavonoids, and by Maillard reaction products.<sup>14,34</sup> When solubilized in water, jaggery yielded solutions with a marked brown colour (see the visual appearance in Table 2), which could negatively affect the characteristics of the impregnated product and its acceptability. The measurements of optical properties revealed that the addition of granulated jaggery to the impregnation solution significantly decreased the  $L^*$  coordinate and that this was more determined by the presence of jaggery than by the percentage of replacement. Similarly, an increase in the  $a^*$  and  $b^*$  coordinates was evident when jaggery was included in the solution. Hue ( $h_{ab}^*$ ) values for jaggery solutions were between red and orange, as opposed to sucrose solution, which was greenish. Chroma or colour saturation ( $C_{ab}^*$ ) intensified with jaggery addition, as did the calculated colour difference ( $\Delta E$ ) with respect to no jaggery addition. These results agree with those reported by Cervera-Chiner *et al.* (2021)<sup>35</sup> for the optical properties of kiwi fruit and strawberry jams formulated with different percentages of jaggery.

### Properties of the VI and OD apple rings

**Characterization of the VI samples.** The properties of vacuum-impregnated apple rings are presented in Table 3. As the impregnation solutions were isotonic with the apple tissue, the water activity for all samples was similar in the range of fresh apples. Moisture content decreased slightly after VI, which could be explained by the loss of native liquid during the vacuum pulse.<sup>13</sup> In the VI process, the introduction of an external solution into the porous structure of the product can

involve the replacement of a part of the product's native liquid. When this occurs, the overall inflow of the external solution, quantified through  $X$ , accounts for the amount of native liquid replaced by the solution.<sup>36</sup> The slight reduction in moisture content could also be due to the increase in the solutes due to sugar impregnation. Remarkable changes were observed in the soluble solid content measured by the Brix degrees, which exhibited a significant increase after VI due to sugars being introduced into the sample with the impregnation solution. The higher solute increase corresponded to apples impregnated with the white sugar solution, with no jaggery replacement as a result of its better impregnation properties.

Regarding optical properties (Table 3), the replacement of air by the impregnation solutions yielded more translucent samples, which is reflected in the lower  $L^*$  values. The increased translucence in VI apple slices, resulting in lower  $L^*$  values, is a known phenomenon that occurs as a result of gas replacement by the impregnation liquid, and the consequent porosity decrease. In line with the results obtained in the present work, Contreras *et al.* reported a decrease in  $L^*$  from 71.8 (1.1) to 41.5 (2.6) in VI apple slices.<sup>37</sup> Among the impregnated samples, increasing the jaggery proportion implied a higher  $L^*$ . Although this could be unexpected due to the darker colour of jaggery solutions, it could be explained by a less successful impregnation of jaggery-containing solutions (Table 2). For the other coordinates,  $a^*$  values moved from green ( $a^* < 0$ ) to red values ( $a^* > 0$ ) when jaggery was added to the impregnation solution. In contrast, changes in  $b^*$  were more marked in samples impregnated with the solution prepared with white sugar. This result aligns with previous findings in sucrose-impregnated Granny Smith apple slices.<sup>37</sup> Colour differences with respect to the fresh apple were higher for the white sugar-impregnated samples, which was consistent with the differences in the  $L^*$  values. The visual appearance of the impregnated samples is illustrated in Fig. 1.

Regarding antioxidant properties, VI did not significantly increase the antioxidant properties of apple rings; in fact, a decrease was observed in some cases (Table 3). One possible

**Table 3** Physicochemical and optical properties of the VI apple rings with isotonic solutions formulated with granulated jaggery (GJ) replacing white sugar in different proportions (0–100% GJ). Average  $\pm$  standard deviation of three replicates<sup>a</sup>

	Fresh	VI0% GJ	VI25% GJ	VI50% GJ	VI100% GJ
$a_w$	0.9888 (0.0008) <sup>b</sup>	0.987 (0.0012) <sup>ab</sup>	0.985 (0.002) <sup>a</sup>	0.988 (0.0013) <sup>b</sup>	0.9879 (0.0003) <sup>b</sup>
$x_w$	0.858 (0.005) <sup>c</sup>	0.828 (0.011) <sup>a</sup>	0.831 (0.004) <sup>ab</sup>	0.84 (0.02) <sup>bc</sup>	0.830 (0.012) <sup>ab</sup>
$^{\circ}\text{Brix}$	10.2 (0.2) <sup>a</sup>	16.3 (0.4) <sup>c</sup>	14.2 (0.2) <sup>b</sup>	14.1 (0.4) <sup>b</sup>	13.9 (0.2) <sup>b</sup>
$L^*$	74 (2) <sup>c</sup>	36 (1.5) <sup>a</sup>	49 (7) <sup>c</sup>	42 (6) <sup>b</sup>	54 (3) <sup>d</sup>
$a^*$	−0.1 (0.9) <sup>b</sup>	−0.7 (0.4) <sup>a</sup>	0.3 (0.3) <sup>b</sup>	0.5 (0.7) <sup>b</sup>	0.5 (0.7) <sup>b</sup>
$b^*$	22 (3) <sup>d</sup>	11 (2) <sup>a</sup>	20 (4) <sup>c</sup>	16 (2) <sup>b</sup>	23(2) <sup>d</sup>
$h_{ab}^*$	90 (2) <sup>b</sup>	94 (2) <sup>c</sup>	89 (1.0) <sup>ab</sup>	88 (3) <sup>a</sup>	89 (2) <sup>ab</sup>
$C_{ab}^*$	23 (3) <sup>d</sup>	11 (1.6) <sup>a</sup>	20 (4) <sup>c</sup>	16 (2) <sup>b</sup>	23 (2) <sup>d</sup>
$\Delta E$	—	40 (2) <sup>d</sup>	26 (8) <sup>b</sup>	33 (7) <sup>c</sup>	21 (4) <sup>a</sup>
Total phenols (mg GAE per g <sub>fp</sub> )	1.09 (0.06) <sup>bc</sup>	1.11 (0.02) <sup>bc</sup>	0.98 (0.04) <sup>a</sup>	1.0 (0.11) <sup>ab</sup>	1.16 (0.07) <sup>c</sup>
Total flavonoids (mg QE per g <sub>fp</sub> )	0.56 (0.03) <sup>ab</sup>	0.521 (0.0010) <sup>a</sup>	0.58 (0.02) <sup>b</sup>	0.643 (0.002) <sup>c</sup>	0.783 (0.012) <sup>d</sup>
DPPH AO activity (mg TE per g <sub>fp</sub> )	0.118 (0.008) <sup>bc</sup>	0.12 (0.018) <sup>c</sup>	0.105 (0.007) <sup>ab</sup>	0.103 (0.008) <sup>a</sup>	0.12 (0.013) <sup>c</sup>
ABTS AO activity (mg TE per g <sub>fp</sub> )	8.6 (0.3) <sup>c</sup>	5.8 (0.3) <sup>b</sup>	4.8 (0.2) <sup>a</sup>	4.9 (0.5) <sup>a</sup>	6.2 (0.6) <sup>b</sup>

<sup>a</sup> a, b, c, d different letters in the same row indicate statistically significant differences at the 95% level ( $p$ -value  $\leq 0.05$ ).



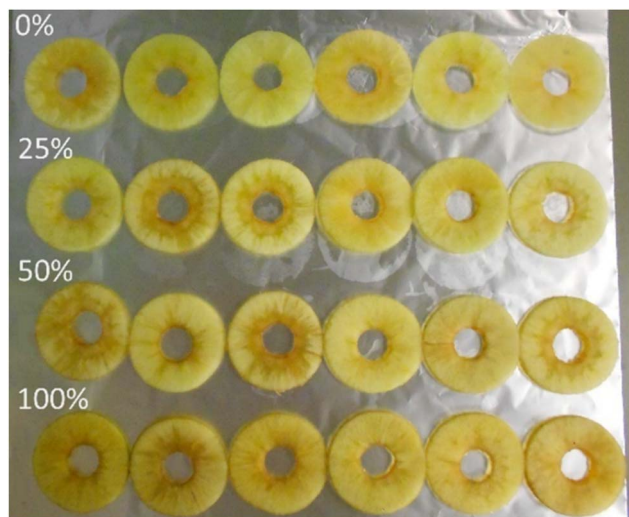


Fig. 1 Apple rings that are vacuum impregnated with isotonic solutions and formulated with white sugar and jaggery. Percentages (0 to 100%) indicate the amount of white sugar replaced by jaggery.

reason for this is the loss of native bioactive constituents, which may occur during VI due to native liquid losses.<sup>38</sup> Nevertheless, the total replacement of white sugar with jaggery improved the antioxidant properties of the samples. In this case, the loss of bioactive compounds due to native liquid outflow could have been compensated for by the antioxidant compounds incorporated with jaggery. In contrast, the 25% GJ solution implied a decrease in all the antioxidant properties evaluated, except flavonoids. Total flavonoid content was the only antioxidant parameter that increased in all cases when jaggery was added to the impregnation solution, as flavonoids are among the major phenolic constituents of sugarcane and its derivatives.<sup>14,39</sup> Antioxidant activities measured using the DPPH and ABTS methods did not improve in the VI samples. The DPPH inhibition in the VI100% GJ samples was similar to that in the fresh apples. In contrast, ABTS antiradical activity decreased in all cases. One possible reason for this is that antioxidant compounds naturally present in apples could be more sensitive

to this method than jaggery constituents. In addition, the shorter reaction times used in the ABTS assay could have contributed to these differences. DPPH radical scavenging activity and total phenolic content obtained for the fresh apple tissue in the present study were slightly lower than those reported by Vega-Gálvez *et al.* for the same apple variety ( $0.266 \pm 0.004$  mg TE per g<sub>fp</sub> and  $1.583 \pm 0.007$  mg GAE per g<sub>fp</sub>, respectively).<sup>40</sup>

**Characterization of OD samples.** Because VI did not produce a significant improvement in antioxidant properties, OD was used as an alternative or complementary treatment to VI, as detailed in the materials and methods section. OD implied a reduction in moisture content in all cases, with no significant differences among the treatments applied; thus, moisture content was reduced from 0.83–0.86 (Table 3) to 0.69–0.71 (Table 4).

In this case, the success of OD treatment for solutes incorporation was evaluated by determining total phenols and flavonoids (Table 4). Combining VI with a subsequent OD treatment did not improve the incorporation of antioxidants into the fruit matrix compared to osmotically dehydrating fresh tissue. Compared to the VI samples, OD succeeded in improving the antioxidant properties of the sample when progressive dehydration was used (pOD), which occurred for both fresh and previous VI apples. This difference between pOD and direct OD could be explained by the fact that direct dehydration in highly concentrated solutions may cause collapsing of the cells next to the osmotic solution, resulting in case hardening and increased resistance to mass transfer in the product surface.<sup>41</sup> Contrarily, when osmotic gradients are progressively applied, water and solute diffusion might be facilitated because this layer of increased resistance is avoided. Besides, dehydration through the tissue symplast is partially preserved because its integrity is better maintained with progressive dehydration.<sup>42,43</sup> These phenomena promote the migration of water from the inner part of the tissue, and a simultaneous counter-current transfer of solutes and bioactive compounds from the osmotic solution to the sample. In the osmotic solution, decreased resistance to mass transfer can also be achieved by modifying the particle size. Filtration of the juice

Table 4 Antioxidant properties of fresh and vacuum-impregnated apple rings. Values are given per gram of fresh product (g<sub>fp</sub>). Average  $\pm$  standard deviation<sup>a</sup>

	VI100% GJ		Fresh	
	OD	pOD	OD	pOD
$x_w$	0.691 (0.003) <sup>a</sup>	0.71 (0.02) <sup>a</sup>	0.700 (0.008) <sup>a</sup>	0.71 (0.013) <sup>a</sup>
Total phenols (mg GAE per g <sub>fp</sub> )	1.07 (0.04) <sup>a</sup>	1.18 (0.08) <sup>ab</sup>	1.09 (0.04) <sup>a</sup>	1.2 (0.2) <sup>b</sup>
Total flavonoids (mg QE per g <sub>fp</sub> )	0.698 (0.009) <sup>a</sup>	0.73 (0.012) <sup>b</sup>	0.70 (0.009) <sup>a</sup>	0.72 (0.02) <sup>b</sup>
$L^*$	44.7 (0.7) <sup>a</sup>	48 (5) <sup>b</sup>	60 (5) <sup>c</sup>	63 (4) <sup>c</sup>
$a^*$	4.4 (0.5) <sup>c</sup>	2 (1.2) <sup>b</sup>	3 (2) <sup>bc</sup>	3 (2) <sup>bc</sup>
$b^*$	29 (1.0) <sup>b</sup>	25 (3) <sup>a</sup>	28 (3) <sup>b</sup>	33 (1.7) <sup>c</sup>
$h$	81 (1.0) <sup>a</sup>	85 (2) <sup>b</sup>	84 (3) <sup>b</sup>	85 (3) <sup>b</sup>
$C$	29 (1.0) <sup>b</sup>	25 (3) <sup>a</sup>	30 (2) <sup>c</sup>	33 (1.9) <sup>c</sup>
$\Delta E$	31.1 (0.6) <sup>c</sup>	27 (5) <sup>b</sup>	18 (4) <sup>b</sup>	16 (4) <sup>a</sup>

<sup>a</sup> a, b, c different letters in the same row indicate statistically significant differences at the 95% level ( $p$ -value  $\leq 0.05$ ).



used as the osmotic agent has been proven to successfully increase antioxidant infusion into the apple tissue during OD as a result of particle size reduction.<sup>44</sup> High-pressure homogenization has been reported to be an interesting technique for reducing particle size in fruit juices and stabilizing bioactive compounds, with a positive impact on impregnation properties.<sup>18</sup> Combining both approaches, *i.e.* particle size reduction and progressive dehydration, or VI, could result in a summative effect.

In Table 3, phenols and flavonoid contents are given per gram of fresh product. Considering moisture content reduction between fresh and osmotically dehydrated samples, OD products (Table 4) exhibited better antioxidant properties per gram of consumed products than fresh and VI products (Table 3). In any case, apples subjected to pOD presented the best antioxidant properties among OD ones. Regarding optical properties, VI samples with further OD were characterized by lower luminosity values. In contrast, the OD applied to the fresh tissue implied a luminosity closer to the fresh tissue. The same was applied to the other colour attributes and colour differences, which were more significant when VI was followed by osmotic dehydration (both OD and pOD) than when OD was used alone. As previously discussed, VI decreases luminosity as porosity is reduced, which implies more significant colour differences. However, the increase in the  $a^*$  coordinate could be due to the inflow of jaggery in the fruit tissue.

**Characterization of HAD and FD apple snacks.** Apple snacks were obtained by applying HAD and FD, as previously explained. Samples subjected to dehydration were fresh apples, VI apples, and pOD apples. Among OD products, the latter was selected because it required a simpler process to obtain a product with a similar or higher antioxidant content than that obtained by VI or a combination of both techniques. The results for snacks, given per gram of dried product, are shown in Table 5. Drying allowed the reduction of water activity values to a range in which stability is significantly increased compared to the fresh apple, thus lengthening the shelf life of the products. In general, FD snacks showed  $a_w$  values smaller than HAD ones. For both HAD and FD snacks, those subjected to previous VI or OD exhibited higher water activity probably due to sugars contributing to water retention. The moisture content of FD samples was also generally lower than HAD ones, except for osmotically dehydrated snacks. FD is characterized by yielding products of very low moisture content (<2–3%) but is usually more hygroscopic due to its characteristic porosity.<sup>45</sup> This, together with the hygroscopic effects of low molecular weight sugars present in jaggery,<sup>46</sup> could be responsible for the increased moisture content of FDpOD samples.

The drying technique used, HAD or FD, had different effects on the properties of VI and pOD jaggery-formulated snacks. When HAD was applied, the use of jaggery in the formulation of apple snacks resulted in a significant improvement in

**Table 5** Antioxidant properties of hot air-dried (HAD) and freeze-dried (FD) apple snacks subjected to previous VI or OD treatments with jaggery solutions. Values are given per gram of dried product ( $g_{dp}$ ). Average  $\pm$  standard deviation of three replicates<sup>a</sup>

Hot air-dried apple snacks	HAD	HAD VI0%	HAD VI25%	HAD VI50%	HAD VI100%	HAD pOD
$a_w$	0.30 (0.02) <sup>a</sup>	0.340 (0.004) <sup>b</sup>	0.34 (0.014) <sup>b</sup>	0.339 (0.009) <sup>b</sup>	0.349 (0.008) <sup>b</sup>	0.348 (0.002) <sup>b</sup>
$x_w$	0.014 (0.005) <sup>a</sup>	0.027(0.006) <sup>b</sup>	0.036 (0.004) <sup>bc</sup>	0.04 (0.010) <sup>c</sup>	0.058 (0.007) <sup>d</sup>	0.074 (0.009) <sup>e</sup>
Total phenols (mg GAE per $g_{dp}$ )	6.3 (0.12) <sup>a</sup>	6 (1.0) <sup>a</sup>	7.4 (0.7) <sup>b</sup>	7.6 (0.8) <sup>b</sup>	8.0 (0.6) <sup>b</sup>	11.0 (0.6) <sup>c</sup>
Total flavonoids (mg QE per $g_{dp}$ )	2.8 (0.10) <sup>a</sup>	3.4 (0.5) <sup>ab</sup>	3.9 (0.7) <sup>bc</sup>	4.1 (0.7) <sup>c</sup>	4.7 (0.6) <sup>d</sup>	7.33 (0.06) <sup>e</sup>
DPPH AO activity (mg TE per $g_{dp}$ )	0.72 (0.03) <sup>d</sup>	0.59 (0.05) <sup>b</sup>	0.7 (0.11) <sup>cd</sup>	0.66 (0.09) <sup>bcd</sup>	0.63 (0.09) <sup>bc</sup>	0.48 (0.03) <sup>a</sup>
ABTS AO activity (mg TE per $g_{dp}$ )	8.1 (0.3) <sup>a</sup>	12.8 (0.8) <sup>b</sup>	16 (2) <sup>c</sup>	15 (1.2) <sup>c</sup>	14.8 (0.8) <sup>c</sup>	23 (1.5) <sup>d</sup>
$L$	74 (2) <sup>c</sup>	55 (3) <sup>a</sup>	53 (6) <sup>a</sup>	56 (4) <sup>a</sup>	55 (5) <sup>a</sup>	62 (2) <sup>b</sup>
$a^*$	8 (1.8) <sup>a</sup>	8 (1.3) <sup>a</sup>	8 (2) <sup>a</sup>	8 (2) <sup>a</sup>	9 (2) <sup>ab</sup>	11 (1.8) <sup>b</sup>
$b^*$	38 (4) <sup>cd</sup>	35 (2) <sup>b</sup>	32 (2) <sup>a</sup>	36 (3) <sup>bc</sup>	38 (1.1) <sup>d</sup>	39 (3) <sup>d</sup>
$h$	78 (2) <sup>b</sup>	77 (1.7) <sup>ab</sup>	76 (4) <sup>ab</sup>	77 (3) <sup>ab</sup>	76 (3) <sup>ab</sup>	75 (1.7) <sup>a</sup>
$C$	39 (4) <sup>cd</sup>	36 (2) <sup>b</sup>	33 (2) <sup>a</sup>	37 (4) <sup>bc</sup>	40 (1.1) <sup>d</sup>	40 (3) <sup>d</sup>
$\Delta E$	—	19 (3) <sup>b</sup>	21 (6) <sup>b</sup>	18 (4) <sup>b</sup>	19 (5) <sup>b</sup>	12 (3) <sup>a</sup>
Freeze-dried apple snacks	FD	FD VI0%	FD VI25%	FD VI50%	FD VI100%	FD pOD
$a_w$	0.190 (0.005) <sup>a</sup>	0.273 (0.006) <sup>bc</sup>	0.266 (0.003) <sup>bc</sup>	0.261 (0.008) <sup>b</sup>	0.261 (0.004) <sup>b</sup>	0.285 (0.028) <sup>c</sup>
$x_w$	0.046 (0.008) <sup>b</sup>	0.03 (0.011) <sup>a</sup>	0.023 (0.005) <sup>a</sup>	0.025 (0.004) <sup>a</sup>	0.02 (0.012) <sup>a</sup>	0.083 (0.006) <sup>c</sup>
Total phenols (mg GAE per $g_{dp}$ )	6.5 (0.2) <sup>d</sup>	4.8 (0.4) <sup>a</sup>	5.2 (0.6) <sup>bc</sup>	5.6 (0.7) <sup>bc</sup>	6.2 (0.9) <sup>cd</sup>	10.8 (0.4) <sup>e</sup>
Total flavonoids (mg QE per $g_{dp}$ )	2.1 (0.12) <sup>a</sup>	2.4 (0.14) <sup>b</sup>	2.7 (0.3) <sup>c</sup>	3.1 (0.2) <sup>d</sup>	3.5 (0.2) <sup>e</sup>	6.93 (0.09) <sup>f</sup>
DPPH AO activity (mg TE per $g_{dp}$ )	0.65 (0.02) <sup>b</sup>	0.47 (0.07) <sup>a</sup>	0.5 (0.10) <sup>a</sup>	0.55 (0.09) <sup>a</sup>	0.6 (0.13) <sup>ab</sup>	0.50 (0.03) <sup>a</sup>
ABTS AO activity (mg TE per $g_{dp}$ )	9.1 (0.2) <sup>a</sup>	10.3 (0.4) <sup>b</sup>	11.3 (0.6) <sup>c</sup>	11.9 (0.4) <sup>c</sup>	13.0 (0.7) <sup>d</sup>	23.8 (0.8) <sup>e</sup>
$L$	86.1 (0.8) <sup>f</sup>	83 (1.5) <sup>e</sup>	81 (1.1) <sup>d</sup>	77 (3) <sup>c</sup>	74 (1.1) <sup>b</sup>	67.2 (0.8) <sup>a</sup>
$a^*$	−1 (1.1) <sup>a</sup>	1 (2) <sup>b</sup>	3 (1.8) <sup>c</sup>	4 (1.5) <sup>c</sup>	5.8 (0.8) <sup>d</sup>	7 (1) <sup>e</sup>
$b^*$	21 (1.4) <sup>a</sup>	26 (2) <sup>b</sup>	28 (3) <sup>bc</sup>	29 (1.8) <sup>cd</sup>	30.3 (0.5) <sup>d</sup>	38 (3) <sup>e</sup>
$h$	92 (3) <sup>d</sup>	88 (5) <sup>c</sup>	85 (2) <sup>b</sup>	82 (3) <sup>b</sup>	79 (1.6) <sup>a</sup>	79.0 (0.9) <sup>a</sup>
$C$	21 (1.4) <sup>a</sup>	26 (2) <sup>b</sup>	28 (3) <sup>bc</sup>	29 (1.9) <sup>cd</sup>	30.9 (0.5) <sup>d</sup>	38 (3) <sup>e</sup>
$\Delta E$	—	6 (2) <sup>a</sup>	9 (3) <sup>b</sup>	13 (3) <sup>c</sup>	16 (1.1) <sup>d</sup>	26 (2) <sup>e</sup>

<sup>a</sup> a, b, c different letters in the same row indicate statistically significant differences at the 95% level ( $p$ -value  $\leq$  0.05).





antioxidant properties in all cases. This was evidenced in all the antioxidant parameters assayed. Both phenols and flavonoids increased, especially flavonoids. For antioxidant capacity, ABTS was significantly more sensitive in this case. Among the air-dried VI and OD samples, phenolic constituents increased as the jaggery concentration increased in the impregnation solution; however, this was not observed when comparing VI samples prior to drying (Table 3). This difference suggests a significant effect of HAD on the antioxidant compounds incorporated through VI and OD. It has been previously reported that thermal treatments may improve the antioxidant properties of foods by promoting biochemical reactions that generate new compounds or isomerization reactions that generate more active forms, either by enzyme activation or inactivation.<sup>47,48</sup> In particular, for sugarcane products and derivatives, previous research<sup>14</sup> showed that moderate thermal treatments imply an increase in the antioxidant capacity of these ingredients. Apple snacks, which were pOD, exhibited the best antioxidant properties. This evidences the capacity of pOD to introduce jaggery bioactive constituents in the apple tissue; additionally, this suggests an ability of OD to better protect native or added bioactive constituents, an effect that has been previously attributed to OD pretreatments.<sup>49</sup>

The addition of granulated jaggery to the fruit matrix by VI or OD did not always imply an improvement in the antioxidant properties of FD snacks, in contrast to HAD snacks. For total phenolic content, improvement was observed only in samples previously subjected to pOD. However, the total flavonoids exhibited a significant increase in FDVI samples when the jaggery concentration in the impregnation solution was higher, thus representing the abundance of these compounds in jaggery. Results obtained for FD samples agree with those obtained for previous VI in which the use of impregnation solutions of low jaggery content does not compensate for the loss of native antioxidants. In the case of HAD snacks, the FD product that presented the best antioxidant characteristics was subjected to pOD, in which all antioxidant attributes improved significantly. The results for FDpOD apple snacks quite agreed with HADpOD ones, thus reinforcing the idea that OD allows the successful introduction of new antioxidants in the fruit matrix and helps protect native or added antioxidant compounds during further processing.<sup>49</sup> DPPH and ABTS antioxidant activities of FD products

followed a similar trend to HAD ones although values were generally lower except for pOD ones. The lower temperatures and vacuum conditions used during FD could have better protected temperature-sensitive antioxidant compounds but did not promote the formation of new or more active compounds or influenced enzymatic reactions, as suggested for HAD. Dalmau *et al.* also reported a significant increase in the antioxidant properties of HAD apple slabs, whereas FD samples exhibited reduced total phenolics and ABTS antioxidant activity compared to the fresh tissue.<sup>50</sup>

The optical properties of apple snacks are presented in Table 4. As in previous analyses, the  $L^*$  coordinate was the most affected one. This was especially evidenced in the HADVI samples, which also presented the highest colour differences with respect to HAD ones, due mainly to differences in luminosity. On the contrary, the  $a^*$  and  $b^*$  parameters were quite homogeneous among the HAD samples. Compared to the non-dried samples (Table 3),  $a^*$  and  $b^*$  values increased significantly after HAD. These changes in the colour coordinates are due to structural and biochemical changes experienced during air drying, such as the contraction of the apple tissue, the concentration of compounds that are responsible for colour (native and incorporated ones), and browning reactions. Similar colour changes have been previously reported for apple slices after convective drying.<sup>37</sup>

Colour differences among VI samples were reduced after FD and colorimetric properties were quite similar for all FD snacks. Luminosity values slightly increased as the proportion of jaggery incorporated into the product increased, but the differences were not as remarkable as before FD. Therefore,  $a^*$  and  $b^*$  were coordinated although less significantly than in HAD snacks. This is due to the use of low temperatures during the process and the reduced exposure to oxygen during sublimation. Among FD samples, colour differences for the FDpOD samples were significantly higher, thus evidencing the incorporation of jaggery-coloured compounds into apple tissue owing to progressive dehydration. Fig. 2 shows the visual appearance of the pOD samples before and after drying by HAD and FD.

**Mechanical properties of selected apple products.** The mechanical properties of the intermediate and final products are presented in this section. Vacuum impregnation with 100% jaggery solution (VI100%) and progressive osmotic dehydration



Fig. 2 Osmotically dehydrated apple rings subjected to progressive osmotic dehydration (pOD). Hot air-dried apple snacks previously subjected to pOD (HADpOD), and freeze-dried apple snacks previously subjected to pOD (FDpOD).





(pOD) was selected for this test. In Fig. 3, force–deformation curves obtained through the puncture test for fresh, VI and pOD, and corresponding FD and HAD samples, are presented.

The curves for fresh and vacuum-impregnated samples were quite similar, presenting multiple fractures after some deformations, with low penetration force and a flat peak typical of fresh porous plant tissues<sup>51</sup> made up of strongly associated turgent cells. In the pOD samples, deformation prior to rupture prolonged significantly, evidencing a more viscoelastic behaviour due to the loss of cellular turgor. However, dehydrated samples exhibited different behaviours depending on the treatment undergone. Higher and sharper peaks were obtained for HAD, which showed a marked increase in force with deformation, indicating increased tissue compactness. Moreover, FD implied an increased force with deformation but was less marked than HAD. This could be due to the porous structure created in the FD samples after sublimation.<sup>52</sup> The trends depicted in Fig. 3 are statistically compared to those in Table 6, where maximum force ( $F_{\max}$ ) and deformation at maximum force ( $d_{\max}/l$ ) values are presented. The maximum force required to penetrate fresh apples is reported as  $7.8 \pm 0.9$  N.<sup>37</sup> After vacuum impregnation (VI), followed by air drying at 30 and 50 °C, this force increased to 20.1 and 18.8 N, respectively, which is consistent with the present findings.

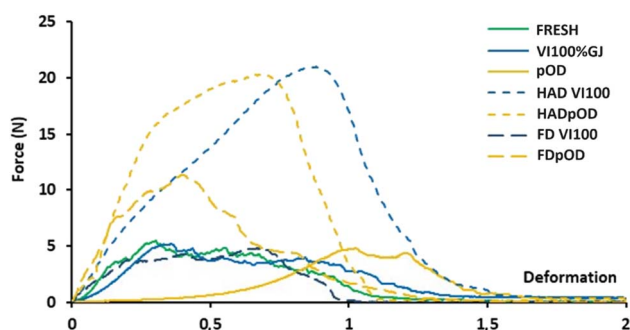


Fig. 3 Force–deformation curves of the puncture test for intermediate and final products, where deformation is obtained as distance/thickness: fresh, vacuum impregnated with 100% jaggery (VI100% GJ), progressive osmotically dehydrated (pOD), hot air- and freeze-dried VI snacks (HAD VI100 and FD VI100), and hot air- and freeze-dried progressively osmotically dehydrated snacks (HADpOD and FDpOD).

Table 6 Mechanical properties of intermediate and final products obtained by puncture test. Maximum force ( $F_{\max}$ , N) and deformation at maximum force obtained ( $d_{\max}/l$ ;  $l$  is the sample thickness)<sup>a</sup>

	$F_{\max}$ (N)	Deformation ( $d_{\max}/l$ )
Fresh	5.4 (1.2) <sup>a</sup>	0.28 (0.04) <sup>a</sup>
IV100%	5.3 (0.7) <sup>a</sup>	0.37 (0.03) <sup>a</sup>
pOD	4.9 (1.0) <sup>a</sup>	0.94 (0.09) <sup>c</sup>
HAD IV100%	21.4 (5.5) <sup>c</sup>	1.4 (0.4) <sup>d</sup>
HAD pOD	18.8 (2.7) <sup>c</sup>	0.58 (0.10) <sup>b</sup>
FD IV100%	5.7 (1.2) <sup>a</sup>	0.48 (0.13) <sup>ab</sup>
FD pOD	11.3 (1.9) <sup>b</sup>	0.44 (0.09) <sup>ab</sup>

<sup>a</sup> a, b, c different letters in the same column indicate statistically significant differences at the 95% level ( $p$ -value  $\leq 0.05$ ).

## Conclusion

The present work explored jaggery as a non-conventional osmotic agent to obtain nutrient rich apple snacks through vacuum impregnation and osmotic dehydration, followed by air-drying or freeze-drying stabilization. The results confirmed that the antioxidant properties of non-centrifugal sugar may be incorporated into the tissue, thus producing fortified apple snacks, especially when progressive osmotic dehydration was applied. Freeze drying helped preserve the antioxidants present in the jaggery-enriched samples, whereas convective hot air drying intensified the antioxidant properties of the jaggery-enriched products. Optical and textural properties were affected by the addition of jaggery and processing techniques. Further studies should explore the acceptability of the products by consumers, as well as the stability of intermediate products and snacks. In conclusion, jaggery is proposed as a healthier osmotic agent to produce more nutritious and sustainable apple snacks using matrix engineering techniques.

## Author contributions

Cristina Barrera: conceptualization, investigation, data curation, formal analysis, methodology, writing – review & editing, supervision, funding acquisition. Noelia Betoret: conceptualization, methodology, writing – review & editing. Lucía Seguí: conceptualization, investigation, data curation, formal analysis, methodology, writing – original draft, writing – review & editing, supervision, project administration, funding acquisition.

## Conflicts of interest

Authors declare no conflict of interest.

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