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ORIGINAL RESEARCH

# Production of Wheat Flour/PBAT Active Films Incorporated with Oregano Oil Microparticles and Its Application in Fresh Pastry Conservation

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**Abstract**

This study aimed to produce wheat flour and poly (butylene co-terephthalate adipate) films with the addition of free and microencapsulated oregano essential oil by blown extrusion and to evaluate its applicability as an active packaging in the preservation of the Brazilian fresh pastry, known as Pastel. The oregano essential oil microparticles were produced by spray drying and characterized in terms of morphology, encapsulation efficiency, and mean diameter. In the films, the physical, antioxidant, and diffusion kinetics of oregano essential oil were determined. Films with microencapsulated oregano essential oil showed lower tensile strength and Young's modulus and higher elongation, solubility, antioxidant capacity, and diffusion coefficient when compared to the control and free oregano oil films. During 28 days of refrigerated storage, fresh pastry packaged with film with oregano essential oil microparticles presented a lower count of molds and yeasts, compared to pastries packed with control and free oregano oil films. The microencapsulation protected oregano essential oil from extrusion condition shearing and temperature damages during film production, allowing the development of biodegradable active films for application in foods where contamination by molds and yeasts is predominant.

**Keywords** Spray drying · Bioactive compounds · Blown extrusion · Active packaging · Diffusion · Shelf life

## Introduction

Wheat is a commodity of great importance to the world economy because of the high consumption of its derivatives. However, a large number of wheat grains are lost or harvested with lower technological quality due to pest attack, germination on the field, or even due to rainfall during harvesting, as well as reduced dry matter as a result of harvest delays. Thus, wheat flour extracted from grains which presents limitations for food applications can be an alternative for the production of biodegradable films. The use of wheat flours to obtain bioplastics is an energetically and economically cheap alternative to purified starch, giving materials interesting functional properties, except for lower strain to break (Benincasa et al., 2017; Leblanc et al., 2008). In this context, the functional and bioactive properties of wheat flour-based films can be improved by blending with other biodegradable polymers, such as poly (butylene co-terephthalate adipate) (PBAT), and by adding bioactive compounds to obtain active packaging (Andrade-Molina et al., 2013; de Campos et al., 2019; de Medeiros et al., 2019; Mücke et al., 2021). PBAT is a biodegradable aliphatic–aromatic co-polyester with tensile properties compared to low-density polyethylene (LDPE) and is

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permitted for food packaging applications by the Food and Drug Administration (Zehetmeyer et al., 2016).

Active packaging is capable of changing the conditions of the packaged food to extend its shelf life, improving sensory properties, or even inhibiting the growth of pathogenic and deteriorating microorganisms (Byun et al., 2010). Among the bioactive compounds to be incorporated in the formulation of active films, oregano essential oil (*Origanum vulgare* L.) stands out for its broad spectrum of antimicrobial and antioxidant activity (Ribeiro-Santos et al., 2018; Ruiz-Navajas et al., 2013; Solano & de Gante, 2012; Viuda-Martos et al., 2010). Several studies show its use in the production of films by casting, and few studies report the production of active films by thermoplastic extrusion (de Medeiros et al., 2019; dos Santos Paglione et al., 2019; Pelissari et al., 2009). During the extrusion process, high temperature, pressure, and shear are necessary to melt the polymer, and as a consequence, the incorporated essential oils can volatilize and even degrade. An alternative to preserve the bioactivity of essential oils and allow their incorporation in the extruded film is to perform their microencapsulation, forming a protective barrier (Ribeiro-Santos et al., 2017).

In the previous work, de Medeiros et al. (2019) developed antimicrobial films of cassava starch and PBAT incorporated with oregano essential oil (OEO) microparticles obtained by ionic gelation. However, the microparticles obtained by ionic gelation had an average of 98% humidity, requiring a drying step to incorporate them into the extruded film, unlike the particles produced by spray drying, in which the final product is in powder form. Spray drying microencapsulation is one of the most commonly used processes in the food industry because it is considered an economical, flexible, continuous, and fast process, and it can be performed with easily accessible equipment that results in good quality dry particles (da Costa et al., 2013). This technique allows microencapsulation of products sensitive to heat, such as essential oils, without drastically affecting their functional properties, allowing their incorporation into the formulation of biodegradable films.

Because of its composition and, mainly, its high moisture content, fresh pasta is subject to the development of a wide variety of microorganisms, which can deteriorate it and even pose a risk to public health. Chemical preservatives, mainly organic acids and its salts (sorbate, benzoate, and propionate), are used, in combination with refrigeration because of the lack of thermal treatment before the packaging, to inhibit the microbial growth (Ouattara et al., 2000; Silveira et al., 2007) and to increase the shelf life of fresh dough. Furthermore, the food industry has been looking to minimize the use of synthetic preservatives, since toxicological studies have demonstrated the possibility that these substances might have some toxic effects (Neltner et al., 2013). In this sense, the use of active packaging containing the essential oil is an interesting option instead of the addition of synthetic preservatives.

The objective of this work was to produce and characterize wheat flour and PBAT films incorporated with free and microencapsulated OEO by blow extrusion. The films were applied as active packaging in fresh pastry, and water activity and mold and yeast counts were monitored during refrigerated storage.

## Materials and Methods

### Materials

The microparticles were produced with OEO (Quinari, Brazil), arabic gum (Nexira, Brazil), maltodextrin (Cargill, Brazil), and paprika oleoresin (Citromax, Brazil). PBAT (Basf, Germany), wheat flour (16.5% moisture, 0.65% ash, 11.1% protein, 1.0% lipids, Vilma Alimentos, Brazil), and glycerol (Dinâmica, Brazil) were employed in film production.

### Microencapsulation of OEO by Spray Drying

For the OEO microencapsulation, preliminary tests were carried out to define the concentration of emulsion solids (30%, wt/wt) and the proportion of wall material (Alvim et al., 2016). Initially, arabic gum and maltodextrin (1:1) were dissolved in distilled water; OEO (3% to emulsion solids) and paprika oleoresin (2% in relation to the OEO mass), whose function was to stain the microparticles to facilitate visualization, were added and stirred for 3 min at 15,000 rpm in Ultra-Turrax (IKA, T-18 model, USA). The emulsion prepared was atomized in a spray dryer chamber through a double fluid atomizer nozzle with 0.7 mm of diameter, an inlet air temperature of 130°C, outlet temperature of 100°C, feed flow of 600 mL $h^{-1}$, airflow of 1.65 $m^3$ $min^{-1}$, and compressed air pressure of 6 bar.

#### Characterization of the OEO Microparticles

The morphology of the microparticles was evaluated by scanning electron microscopy (SEM). The sample was covered with gold and then visualized on the SEM (Philips, FEI Quanta 200 model, Japan) with an acceleration voltage of 20 kV. The magnitude of observation was 5000× (Alvim et al., 2016).

The mean diameter ($D_{50}$) and size distribution of the microparticles were determined by light scattering (Horiba, LV950 model, Japan) using absolute ethanol as the dispersing medium (Alvim et al., 2016). The polydispersity was given by the span index (Eq. 1).

$$\mathrm{Span} = (D_{90} - D_{10})/D_{50} \qquad (1)$$

where $D_{10}$, $D_{50}$, and $D_{90}$ are 10%, 50%, and 90% of the cumulative distribution, respectively.

The analysis of the encapsulation efficiency (EE) was performed in triplicate using the steam distillation with a Clevenger. EE was determined considering the relation of the amount of oil extracted from the microparticles and the initial amount of oil (de Medeiros et al., 2019; dos Santos Paglione et al., 2019).

## Production of Wheat Flour and PBAT Films by Blown Extrusion

The formulation of the control film consisted of 46% (wt/wt) wheat flour, 40% (wt/wt) PBAT, and 14% glycerol (wt/wt) (Table 1). The addition of 10% of microparticles in the formulation of the film with OEO microparticles (FOM) was determined according to previous tests, and based on the analysis of microparticles encapsulation efficiency, it was found that in 50 g of microparticles, there were 3.3 g of OEO, and this amount was added to obtain the film with free OEO (FO).

The films were produced according to de Medeiros et al. (2019) with minor modification. The blends were processed in a pilot single-screw extruder (BGM, model EL-25, Brazil), with a screw diameter of 25 mm, a length of 28D, a screw speed of 35 rpm, and a temperature profile of 90/120/120/100°C. The cylindrical profiles were pelletized and extruded again to produce films by blown extrusion under the following conditions: temperature profile of 90/120/120/120/120°C, screw speed of 40 rpm, and a 50-mm film-blowing dye with internal air to form the film balloon (Fig. 1). To avoid the formation of the film balloon with tearing or cracking, the winding speed and airflow in the matrix were adjusted for each formulation.

## Film Characterization

Before the characterization tests, all the films were conditioned at 53% controlled relative humidity for at least 48 h at 25°C.

### SEM

The films were fractured in liquid nitrogen, coated with gold in a Sputter Coater (BAL-TEC, SCD-050 model, Balzers, Liechtenstein), and the surface and fracture area were observed using a scanning electron microscope (Philips, FEI Quanta 200 model, Japan) with an acceleration voltage of 20 kV. The magnitude of observation was 1600× for the fracture and 400× for the surface (de Souza et al., 2020).

### Optical Properties

The color of the films was measured using a colorimeter (Konica Minolta, CR-410 model, Japan) with $D_{65}$ illuminant (daylight). The samples were placed in direct contact with the sensor to measure the color parameters L* luminosity (black/white), a* (green/red), and b* (blue/yellow). Ten random measurements were taken for each formulation. The apparent opacity (Y) was determined in triplicate with the same colorimeter and was calculated according to Eq. 2 (Garcia et al., 2014):

$$Y = (Y_b / Y_w) \times 100 \tag{2}$$

where Y is apparent opacity (%), $Y_b$ is L* measured against a black background, and $Y_w$ is L* measured against a white background.

### Mechanical Properties

The tensile test was performed using a texturometer (Stable Micro Systems, TA-TX2 model, England), and the properties obtained were tensile strength (MPa), elongation at break (%), and Young's modulus (MPa) (ASTM, 2001). Ten samples (50 mm × 20 mm) of each formulation were prepared. The films were fixed to the grips of the equipment with an initial distance of 30 mm and a crosshead speed of 0.8 mm/s.

### Water Vapor Permeability (WVP) and Solubility in Water

The WVP was performed in triplicate according to the American Society for Testing and Material (ASTM, 2000) using a relative humidity gradient of 0–75%.

The solubility in water was determined in triplicate according to Gontard et al. (1994) and was calculated as the percentage of dry matter of the solubilized films (20 × 20 mm) after immersion for 24 h in 200 mL of water at 25°C under mechanical stirring.

**Table 1** Formulation of the wheat flour/PBAT films incorporated with free and encapsulated OEO

| Formulation | Wheat flour (g) | Glycerol (g) | PBAT (g) | Microparticle (g) | OEO (g) |
|---|---|---|---|---|---|
| FC | 230 | 70 | 200 | - | - |
| FOM | 207 | 63 | 180 | 50 | - |
| FO | 207 | 63 | 180 | - | 3.3 |

*FC*, control film; *FOM*, film with OEO microparticles; *FO*, film with free OEO

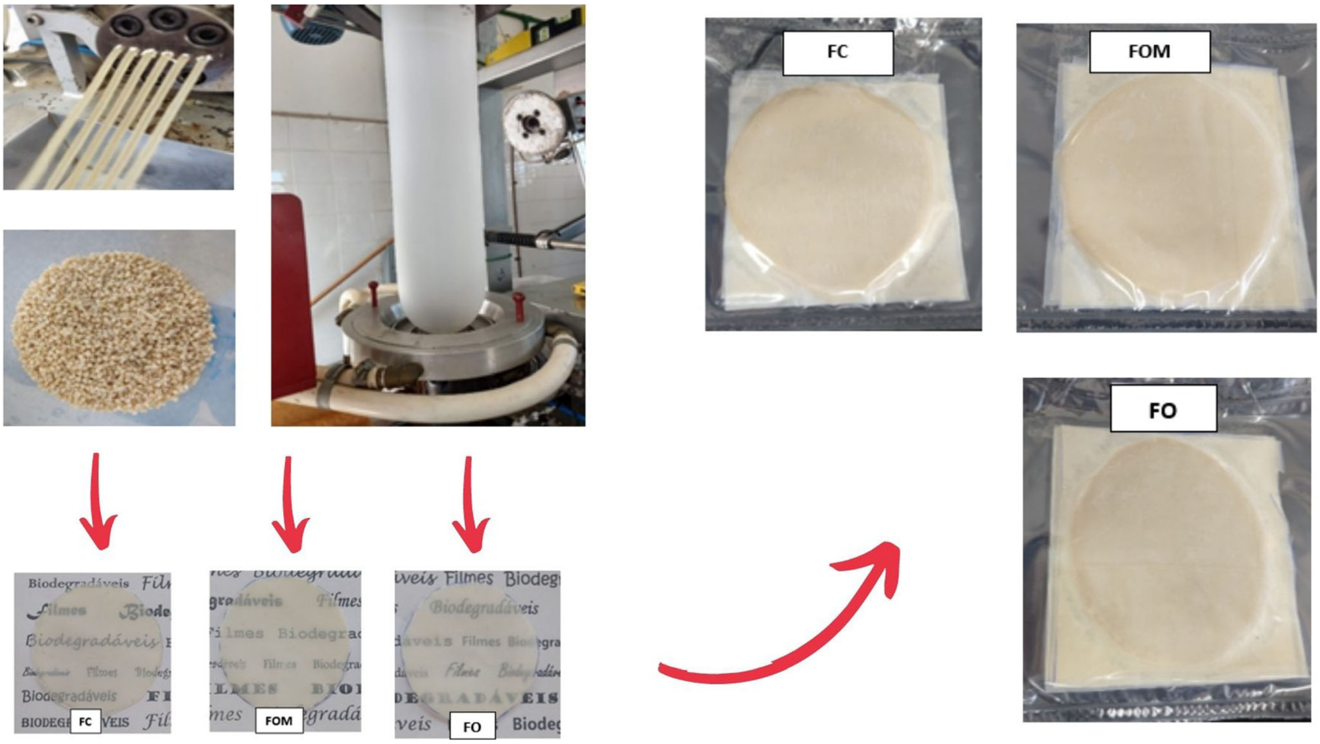

**Fig. 1** Experimental scheme of the production of the wheat flour/PBAT films by blown extrusion and their application in Brazilian fresh pastry (pastel)

### Phenolic Compounds and Antioxidant Capacity

The extraction of phenolic compounds from the films was performed according to previous works (de Souza et al., 2020; dos Santos Paglione et al., 2019). The total phenolic compound was determined according to Folin–Ciocalteu method (Singleton & Rossi, 1965) and expressed as mg EAG/g film. The antioxidant capacity was evaluated by the iron reduction method (ferric reducing ability of plasma, FRAP) (Benzie & Strain, 1996), by capturing the free radical DPPH (Mensor et al., 2001) and ABTS (Thaipong et al., 2006) methods. The results were expressed in μM Trolox equivalent per g of film.

### Release of OEO in Food Simulants

The migration study of OEO was performed according to Ke et al. (2019), using ethanol 10% and 95% (v/v) as aqueous and fat simulant, respectively. In a brown flask, the films (60 mm × 100 mm) were immersed in 50 mL of simulant and kept under agitation (100 rpm) at 25°C. At different times, an aliquot of 2 mL of solution was removed and the concentration of OEO released was quantified by ultraviolet–visible spectroscopy (230 nm). Before testing, Full-Band Scanning was used to determine the maximum absorption peak of OEO, and the standard curve was created.

For the OEO release kinetics, the Power Law model or Korsmeyer–Peppas model was used:

$$\frac{M_t}{M_\infty} = k.t^n \tag{3}$$

where $M_t/M_\infty$ is the amount of OEO released at each time point t relative to the amount released at equilibrium, k is a kinetic constant, and n is the diffusion exponent indicating the type of release mechanism as Fickian diffusion ($n = 0.5$), anomalous diffusion ($0.5 < n < 1.0$), or non-Fickian diffusion ($n = 1.0$). The adjustment was applied to $M_t/M_\infty < 0.6$.

The Weibull model was used to evaluate the diffusion behavior of the OEO for the entire range of $M_t/M_\infty$ measurement:

$$\frac{M_t}{M_\infty} = 1 - \exp\left(\alpha . t^\beta\right) \tag{4}$$

where α and β are constants. The β parameter indicates the release mechanism as Fick's diffusion ($\beta < 0.75$), case II transport ($0.75 < \beta < 1$), and complex release ($\beta > 1$).

The mass diffusion of OEO was modeled with Fick's second law obtaining by the Laplace transform method:

$$\frac{M_t}{M_\infty} = \frac{2}{L}\sqrt{Dt}\left[\frac{1}{\sqrt{\pi}} + 2\Sigma_{n=1}^{\infty}(-1)^n \text{ ierfc } \frac{nL}{\sqrt{Dt}}\right] \tag{5}$$

where $Mt/M_\infty$ is the amount of OEO released at each time point t relative to the amount released at equilibrium, L is half the thickness of the film, D is the diffusion coefficient, and t is the time.

According to the variable separation method, we obtain

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}.\exp\left(-\frac{(2n+1)^2\pi^2}{4L^2}Dt\right) \qquad (6)$$

Equations 5 and 6 can be simplified for short diffusion times, and the series of Eq. 6 tends to zero, obtaining the following equation for $M_t/M_\infty < 0.3–0.5$:

$$\frac{M_t}{M_\infty} = \frac{2}{L}\sqrt{\frac{Dt}{\pi}} \qquad (7)$$

The diffusion coefficient was obtained using the graph composed of the ratio $M_t/M_\infty$ versus $t^{1/2}$.

For long-term diffusion or values of the $M_t/M_\infty > 0.5–0.7$, the first term in the series of Eq. 6 is preponderant, and thus, the series can be truncated in the first term, resulting in

$$\frac{M_\infty - M_t}{M_\infty} = \frac{8}{\pi^2}\exp\left(-\frac{\pi^2 Dt}{4L^2}\right) \qquad (8)$$

The diffusion coefficient was obtained using a graph composed of the terms log $((M_t - M_\infty)/M_\infty)$ versus t.

### Application of the Films in the Brazilian Fresh Pastry (Pastel) Conservation

The films produced were applied in commercial Brazilian fresh pastry without the addition of preservatives. Dough disks with 100 mm of diameter and an average thickness of 1.06 mm were placed on the film, and then another film was placed on the dough. This film and dough intercalation procedure was repeated 3 times, and the whole was packed in polyethylene bags and sealed (Fig. 1). The fresh pastry was stored at 7 ± 1°C for 28 days, and water activity and mold and yeast counting were performed every 7 days until the 28th day of storage.

### Statistical Analysis

The means of the results were evaluated using the analysis of variance (ANOVA), and the means of the treatments were compared using the Tukey test at the level of 5% significance ($p < 0.05$) using the Statistica 12.0 software (Statsoft, USA).

## Results and Discussion

### Characterization of the OEO Microparticles

According to SEM images (Fig. 2), the OEO microparticles showed spherical geometry with the outer surface wrinkled due to the shrinkage of the drops during the drying and cooling steps. The microparticles formed clusters that may contain oil in the interstices, aiding in their retention. The EE of the OEO microparticle was 65.7%. A higher value (86.2%) was obtained by Toledo Hijo et al. (2015), probably due to the use of different wall materials and process conditions.

The mean diameter of the microparticles was 8.32 μm, and the approximate value was observed by other authors encapsulating oregano and rosemary essential oil by spray drying (Teodoro et al., 2014; Toledo Hijo et al., 2015). The span values characterize how homogeneous the particle size distribution is, that is, the higher the span value, the more heterogeneous or polydispersed its distribution is. The span index was 1.58, which is considered high and indicates that there was no homogeneity regarding the size of the samples (Alvim et al., 2016). The high span value corroborates with the SEM image that revealed particles with different sizes and is related to the use of the double fluid nozzle which, during the spraying of the emulsion, does not form drops with uniform size.

### Visual Aspect, Morphology, and Optical Properties

The films presented a slightly yellowish, opaque color, homogeneity, and good handling (Fig. 1). There was no significant difference in thickness between the FOM and FO films, but between these films and the control film (FC), there was a significant difference, as shown in Table 2. In blow extrusion, thickness is mainly controlled by winding speed and airflow in the balloon matrix, and the greater thickness of the FO and FOM films was due to the reduction of the airflow during the formation of the balloon.

According to the film's SEM images (Fig. 2), the surface revealed an increase in roughness because of the addition of free and encapsulated OEO, which can be related to their interference in polymer flow during the extrusion process. The presence of particles of different sizes may be related to non-gelatinized starch and proteins such as gluten and fibers. The fracture images showed an irregular structure due to the presence of several components that compose the blend, and no significant difference was observed between the formulations.

The films did not show a significant difference for L* and b* parameters (Table 2, $p > 0.05$). However, the addition of OEO microparticles contributed to a higher value of a* because of the paprika oleoresin. The L* value of FC was lower than that reported by da Silva et al. (2019) and Mücke et al. (2021) in cassava starch and PBAT films (70:30) produced by blow extrusion (94.84 and 90.21), and this is related to the use of wheat flour. The darker color of the films in this work may be also due to the Maillard reaction between proteins and

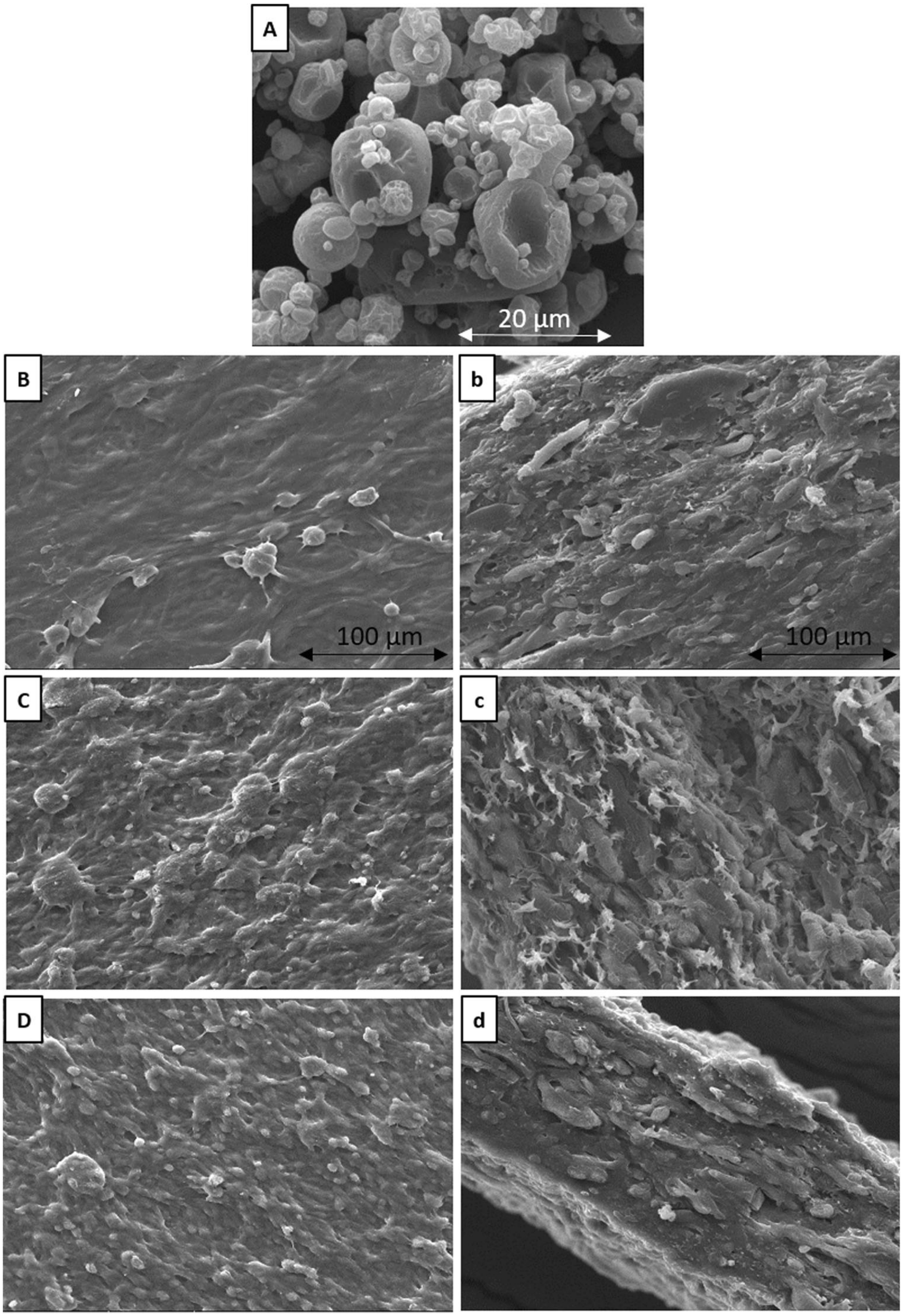

**Fig. 2** SEM image of OEO microparticle and wheat flour/PBAT films. OEO microparticles (A), control film surface (B) and fracture (b), film with OEO microparticle surface (C) and fracture (c), and film with free OEO surface (D) and fracture (d)

**Table 2** Color, opacity, and thickness of wheat flour/PBAT films incorporated with free and encapsulated OEO

| Film | Thickness (μm) | L* | a* | b* | Opacity (%) |
|---|---|---|---|---|---|
| FC | $117 \pm 18^{a}$ | $88.61 \pm 1.10^{a}$ | $0.29 \pm 0.23^{a}$ | $7.48 \pm 2.91^{a}$ | $58.27 \pm 1.81^{a}$ |
| FOM | $170 \pm 38^{b}$ | $88.44 \pm 1.17^{a}$ | $0.57 \pm 0.06^{b}$ | $8.53 \pm 3.56^{a}$ | $61.46 \pm 0.86^{ab}$ |
| FO | $175 \pm 26^{b}$ | $88.03 \pm 1.01^{a}$ | $0.22 \pm 0.09^{a}$ | $9.38 \pm 2.89^{a}$ | $64.19 \pm 1.50^{b}$ |

*FC*, control film; *FOM*, film with OEO microparticles; *FO*, film with free OEO. $^{a,b,c}$ Means followed by different letters in the same column show a significant difference ($p < 0.05$) according to Tukey's test carbohydrates of the wheat flour and the use of high extrusion temperature (130°C).

Regarding the apparent opacity of the films, the addition of free and microencapsulated OEO increased the opacity, and we attributed this phenomenon to the OEO which has a yellowish color that prevents the passage of light through the film or also to the light scattering effect caused by the OEO lipid droplets (Liu et al., 2019).

## Mechanical Properties

The results of the mechanical properties of wheat flour/PBAT films containing free and encapsulated OEO are shown in Table 3. In general, it was found that the FOM showed lower values of the tensile strength (T) and Young's modulus (YM) in relation to the other formulations, and this can be related to the tension concentration points that the microparticles produced. Also, the plasticizing effect of OEO stands out, since there was also a reduction in the YM of the FO and FOM and a significant increase in the elongation of the FOM and FO. The plasticizing effect of OEO was previously verified in films produced by casting and blown extrusion (Benavides et al., 2012; de Medeiros et al., 2019; dos Santos Paglione et al., 2019; Pelissari et al., 2009; Solano & de Gante, 2012).

de Medeiros et al. (2019) also verified, in cassava starch/PBAT films incorporated with OEO microparticles obtained by ionic gelation, a reduction of T and YM for films added with microparticles when compared to the control film. On the other hand, in SPC (soy protein concentrate) films added with OEO microencapsulated by ionic gelation, there was an increase in T and YM, this result being related to the good interaction between SPC and the microparticle sodium alginate that resulted in the reinforcement of the matrix (dos Santos Paglione et al., 2019).

The films of wheat flour and PBAT produced in this work presented superior mechanical properties than extruded films produced only with wheat flour produced by Benincasa et al. (2017) (T ranged from 0.72 to 1.75 MPa and ε of 38.33 to 115.82%) and Puglia et al. (2016) (T of 0.7 to 1.4 MPa and ε of 36 to 72%), with values of T and ε approximately three times greater. This is due to the formation of a blend with PBAT, which has superior mechanical properties, and according to Zhai et al. (2020), when the PBAT content exceeded 30 wt%, a significant improvement of the mechanical strength and flexibility of wheat flour-based films can be obtained. In addition, the improvement of the mechanical properties of the wheat flour-based film could be related to the increased interaction between the starch, protein, and PBAT during thermoplastic extrusion.

## WVP and Water Solubility

The WVP of the films ranged from $1.92 \times 10^{-7}$ to $2.18 \times 10^{-7}$ g m$^{-1}$ h$^{-1}$ Pa$^{-1}$, showing no significant difference ($p > 0.05$) (Table 3) between treatments. This result may be related to the film morphology presented previously (Fig. 1), in which we found that there were no differences in the fracture images. The WVP of the films is influenced by the hydrophilic or hydrophobic nature of the material, the presence of pores and cracks, and tortuosity through the film (Bertuzzi et al., 2007). de Medeiros et al. (2019) found that the WVP of the films containing OEO microcapsules did not differ significantly from the control films and with free oil, as the amount of particle added was 1% in relation to the total mass of the formulation and may not have interfered in the continuous polymer phase. A different behavior was reported by dos Santos Paglione et al. (2019) in which SPC films containing OEO microparticles had lower WVP compared to the control and incorporated with free OEO films, and the authors attributed this to the good interaction between SPC and alginate used as wall material. The WVP values found in this study were close to those reported by da Silva et al. (2019) in thermoplastic starch (TPS), PBAT, and *Pinhão* extract film and by Garcia et al. (2014) in TPS, PBAT, and citric acid films.

In general, the package should possess desirable water resistance and should maintain its integrity during storage. Thus, solubility determination is critical for the characterization of hydrocolloids based films (Mushtaq et al., 2018). The water solubility of the films ranged from 3.71 to 14.30% (Table 2), and the FO showed no significant difference ($p > 0.05$) when compared to FC. The FOM showed a significant increase in solubility values, and this may be related to the presence of arabic gum and maltodextrin (wall materials),

**Table 3** Mechanical properties, water vapor permeability, and solubility of wheat flour/PBAT films incorporated with free and encapsulated OEO

| Film | T (MPa) | ε (%) | YM (MPa) | WVP × $10^7$ (g m$^{-1}$ h$^{-1}$ Pa$^{-1}$) | Solubility (%) |
|---|---|---|---|---|---|
| FC | $3.87 \pm 0.32^{b}$ | $296.67 \pm 46.50^{a}$ | $19.16 \pm 1.13^{c}$ | $1.92 \pm 0.07^{a}$ | $4.25 \pm 1.59^{a}$ |
| FOM | $3.01 \pm 0.12^{a}$ | $351.65 \pm 28.35^{b}$ | $15.20 \pm 0.75^{a}$ | $2.08 \pm 0.13^{a}$ | $14.30 \pm 0.26^{b}$ |
| FO | $3.68 \pm 0.16^{b}$ | $313.67 \pm 52.60^{ab}$ | $17.34 \pm 1.04^{b}$ | $2.18 \pm 0.01^{a}$ | $3.71 \pm 1.25^{a}$ |

*FC*, control film; *FOM*, film with OEO microparticles; *FO*, film with free OEO; *T*, tensile strength; *ε*, elongation at break; *YM*, Young's modulus; *WVP*, water vapor permeability. $^{a,b,c}$ Means followed by different letters in the same column show a significant difference ($p < 0.05$) according to Tukey's test which show hydrophilic character and good solubility in water. The significant effect of the microparticles is highlighted since they corresponded to approximately 10% of the film formulation. The solubility values were lower than that obtained by da Silva et al. (2019) in TPS, PBAT, and *Pinhão* extract film (24.68 to 28.15%) and by Mücke et al. (2021) in TPS, PBAT, and water-soluble curcumin films (20.45 to 24.14%), possibly due to the higher concentration of PBAT (40%) used in the present work that has hydrophobic character.

## Phenolic Compounds and Antioxidant Capacity

The results of the phenolic compounds and antioxidant capacity evaluated by the FRAP, DPPH, and ABTS methods of the films are shown in Fig. 3. For the FC film, the value of 1.29 mg EAG/g of film of total phenolic compounds was obtained, and antioxidant capacity was detected by the three methods studied. This may be related to the presence of phenolic acids such as ferulic, syringic, p-cumaric, vanillic, and caffeine present in wheat flour and with antioxidant capacity (Lv et al., 2012).

The addition of free and microencapsulated OEO significantly increased the concentration of total phenolic compounds and the antioxidant capacity of the films, being in greater proportion in the FOM, showing the efficiency of microencapsulation in preserve the bioactive compounds. The main phenolic compounds present in OEO and responsible for the antioxidant capacity are carvacrol and thymol (Boskabady et al., 2014; Selzer et al., 2013). Several works demonstrated the antioxidant capacity of the OEO incorporated film (Cardoso et al., 2017; dos Santos Paglione et al., 2019; Lee et al., 2016).

## Release of OEO in Food Simulant

Table 4 shows the parameters of the Power Law model and the Weibull model for the OEO release kinetics of FO and FOM films in 10% and 95% ethanol. In general, the two models studied showed good adjustment to the experimental data, with $R^2 > 0.91$, being adequate to predict the release mechanism of the OEO from films. Evaluating the n parameter of the Power Law model for all films, independent of the simulant fluid, the values ranged from 0.7727 to 0.8514, and the release of OEO from the wheat flour/PBAT film is classified as anomalous. In this case, the diffusion speed and mobility of the polymer chain segment are comparable and are dependent on the swelling kinetics of the matrix. These results suggest that the film matrix is predominantly hydrophilic since the largest proportion of the formulation is composed of wheat flour and glycerol (~60%). Thus, when the film comes into contact with water, it swells causing the chains to relax and making the water not so readily available for the release of OEO.

For the Weibull model, the β parameter values ranged from 0.8026 to 0.8673, indicating that the OEO release mechanism from the film for ethanol 10% and 95% follows the case II transport, in which the release is governed by a swelling

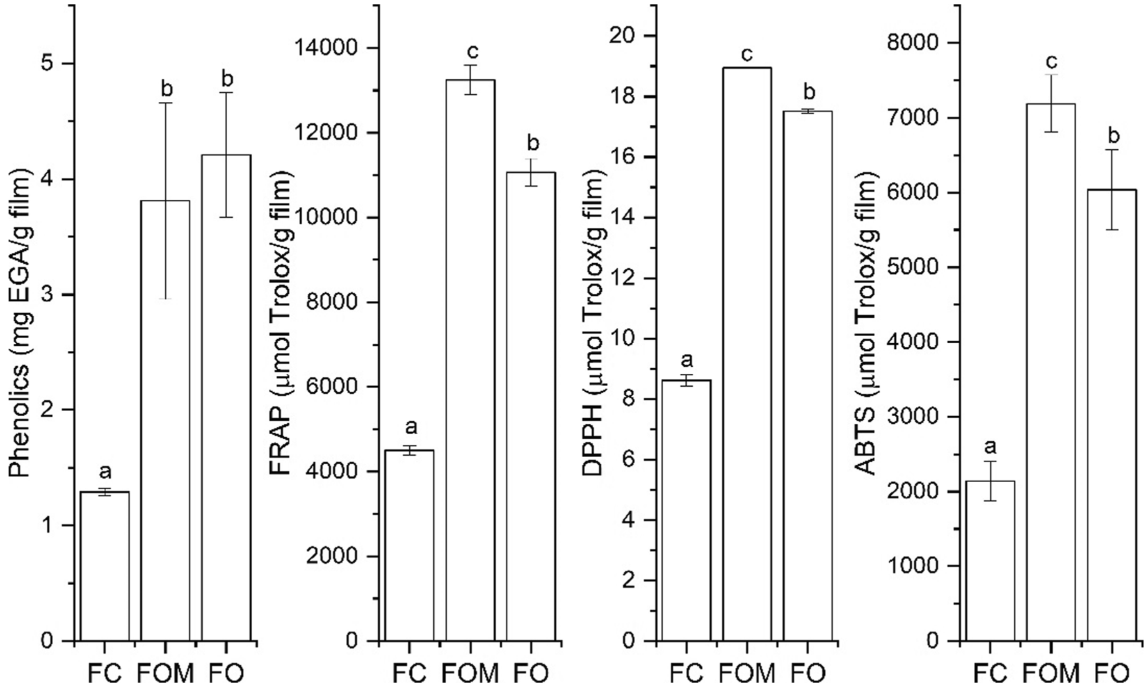

**Fig. 3** Total phenolic compounds and antioxidant capacity evaluated by FRAP, DPPH, and ABTS methods of wheat flour/PBAT films with encapsulated (FOM) and free OEO (FO)

**Table 4** Parameters of Power Law model and Weibull model and diffusion coefficients for short- and long-term migration of OEO from wheat flour/PBAT films in different food simulant

| | Power Law model | | | Weibull model | | | |
|---|---|---|---|---|---|---|---|
| | $n$ | Equation | $R^2$ | $\alpha$ | $\beta$ | Equation | $R^2$ |
| Ethanol 95% | | | | | | | |
| FO | 0.7827 | y = 0.7827x − 10.902 | 0.9140 | $1.63159 \times 10^{-5}$ | 0.8032 | y = 0.8032x − 11.0234 | 0.9515 |
| FOM | 0.7727 | y = 0.7727x − 10.797 | 0.9249 | $1.64793 \times 10^{-5}$ | 0.8026 | y = 0.8026x − 11.0134 | 0.9521 |
| Ethanol 10% | | | | | | | |
| FO | 0.8514 | y = 0.8514x − 11.88 | 0.9260 | $6.41891 \times 10^{-6}$ | 0.8673 | y = 0.8673x − 11.9563 | 0.9501 |
| FOM | 0.8271 | y = 0.8271x − 11.535 | 0.9262 | $8.73064 \times 10^{-6}$ | 0.8464 | y = 0.8464x − 11.6487 | 0.9509 |
| | Short term | | | | Long term | | |
| | $D \times 10^{11}$ (cm²/s) | Equation | | $R^2$ | $D \times 10^{11}$ (cm²/s) | Equation | $R^2$ |
| Ethanol 95% | | | | | | | |
| FO | 1.5329 | y = 0.0007x − 0.0329 | | 0.9868 | 0.5902 | $y = -3.50 \times 10^{-7}x - 0.4088$ | 0.7503 |
| FOM | 2.0388 | y = 0.0007x − 0.033 | | 0.9869 | 0.9740 | $y = -4 \times 10^{-7}x - 0.4162$ | 0.8247 |
| Ethanol 10% | | | | | | | |
| FO | 1.4679 | y = 0.000660022x − 0.03891 | | 0.9887 | 0.7101 | $y = -4.08405 \times 10^{-7}x - 0.53158$ | 0.7888 |
| FOM | 3.6692 | y = 0.000641788x − 0.04002 | | 0.9891 | 1.8624 | $y = -4.05186 \times 10^{-7}x - 0.50222$ | 0.8248 |

*FOM*, film with OEO microparticles; *FO*, film with free OEO

mechanism and relaxation of polymer chains. This result corroborates those observed for the parameters obtained in the Power Law model, suggesting that in a matrix with certain hydrophilicity, such as the films in this work, more than one mechanism is responsible for OEO release.

The two models applied in this work indicated that the OEO release mechanism does not follow Fick's law, as demonstrated by other works that used biopolymers in the film formulation (Ke et al., 2019; Rivero et al., 2013). Among the reasons that explain this difference, it is possible to highlight the blending between wheat flour, which has a hydrophilic character, and PBAT, which has a hydrophobic character; and the difference in the morphology of the films, since the SEM images (Fig. 2) did not reveal a smooth and homogeneous surface and fractures, as observed in films produced by casting. No difference was observed between the release kinetics in the films with free and encapsulated oil, indicating that regardless of their form of addition, OEO has the same release mechanism in the studied media.

Table 4 also shows the diffusion coefficients of OEO in short- and long-term migration in 10% and 95% ethanol. Evaluating the effect of the simulating medium, there was a certain increase in the D values of the FOM when using 10% ethanol, that is, there was greater diffusion of the OEO in this medium. It is possible that in a hydrophilic medium, the water has hydrated and consequently swelled the film matrix, facilitating the diffusion of the OEO.

The diffusion coefficient in long-term diffusion was lower than in short-term diffusion, which could be due to the mass transfer or release, being proportional to the concentration gradient of the system. Therefore, in the initial stage, there is little OEO in the simulant fluid, and this causes a large gradient of concentration between the film and the OEO, leading to a high diffusion rate in the initial stages. During the long-term diffusion stage, the diffusion coefficient decreases due to an increase in OEO in the simulant fluid and a decrease in the concentration of OEO in the film (Ke et al., 2019).

Evaluating the effect of adding free and microencapsulated OEO to the film, it was found that for both 95% and 10%

ethanol, the FOM formulation had a higher D value. Considering FOM had a higher solubility value (Table 2), it is possible that it has a greater hydrophilic character due to the presence of microparticles, which was also produced with water-soluble wall materials, and, therefore, resulted in a greater OEO release. Many factors, such as the physicochemical properties of polymers and active agents, intermolecular interactions, temperature, and solubility, affect the diffusion of the active agent in several phases (Li et al., 2006). These results also confirm the higher values of phenolic compounds and antioxidant capacity of the FOM, indicating that this is an interesting material for packaging foods with a high moisture content such as fresh pastry, in which the release of the bioactive compound is expected as a consequence of hydration and swelling of the matrix.

## Application of Films on Fresh Pastry Conservation

The average Aw (Fig. 4) of the pastry in the initial time was 0.966, a value that favors the development of bacteria and fungi, and therefore, microbiological control through the use of active packaging is an interesting and viable alternative to direct addition of the preservative in the fresh pastry surface, preventing the microorganism's development (Andrade-Molina et al., 2013). In general, for all films, there was a reduction in the Aw values of the pastry during storage due to the migration of the moisture from the pastry to the films. Similar behavior was reported by Andrade-Molina et al. (2013) in fresh pastry intercalated with starch, PBAT, and potassium sorbate films, in which a reduction of 0.04 in Aw was detected after 30 days of storage. On the 28th day of storage, an increase in Aw was observed in the pastry packaged with FC and FOM and may be related to the reverse migration of moisture from the film to the pastry.

The pastry in contact with control and FO films showed an initial count of molds and yeasts of 3.00 log UFC/g and a final count of 3.81 and 3.90 log UFC/g, respectively, with a significant increase during 28 days of refrigerated storage. This result indicates that FO was not efficient in controlling mold and yeast. On the other hand, the pastry in contact with FOM showed a slight increase in count on the 14th day of storage but decreased from the 21st day, remaining similar to the initial count until the end of the storage. This may have occurred due to the slow and gradual migration of the OEO from the film to the food surface, as observed in the OEO release assay discussed earlier. FOM film was more efficient in the control of mold and yeast than FO after 14 days of storage, and this result may also be related to the lower diffusion coefficient of the FO film observed previously.

According to Rivero et al. (2013), the shelf life of the fresh pastry was defined as the time required for the microbial counts (in this case, fungi and yeasts) to reach levels of $10^6$ CFU $g^{-1}$ and absence of pathogenic microorganisms. Considering the authors, the pastry of this work would still be suitable for consumption during the 28 days of storage, regardless of the film used. This fact demonstrates the applicability of biodegradable films produced in the present work, for refrigerated pastry dough conservation, and confirmed that natural preservative such as OEO is efficient in the control of molds and yeasts, extending the shelf life of fresh pastry. Also, it was possible to demonstrate that the incorporation of OEO microparticles in the wheat flour/PBAT film controlled the OEO diffusion, allowing extended release of OEO from film to fresh pastry during refrigerated storage. This is an important information apart of the microorganism's control, considering that, for certain products, a constant diffusion of the active compound, especially essential oils, is more interesting than a rapid diffusion, concerning aroma and sensory issues.

Several researches demonstrate the application of active films in fresh pastry, showing their efficiency in controlling or inhibiting microbial growth. Sousa et al. (2016) proved the efficiency of rice flour, PBAT, and potassium sorbate films in preserving fresh pasta for lasagna. Andrade-Molina et al. (2013) observed an increase in the shelf life of fresh pasta

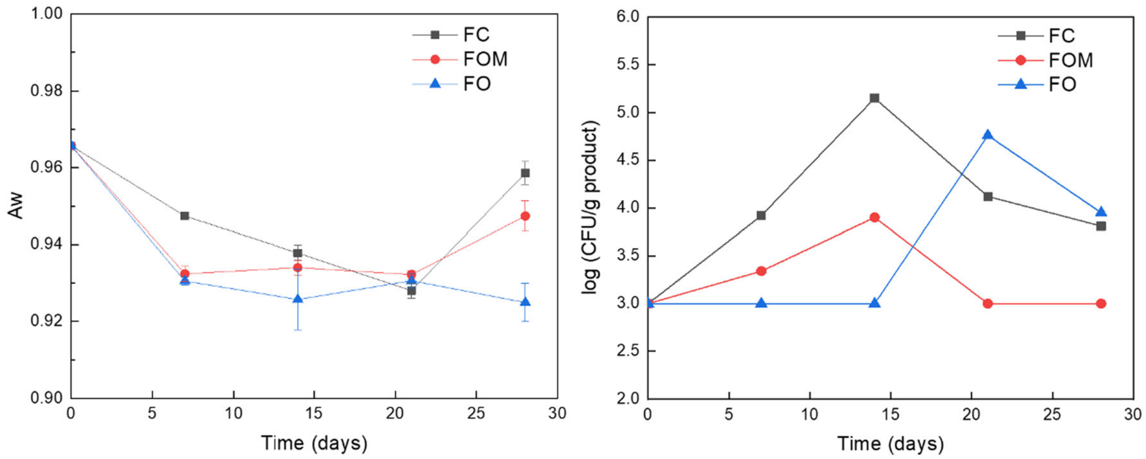

**Fig. 4** Water activity (**a**) and mold and yeast counts (**b**) of the fresh pastry packaged with wheat flour/PBAT films

using starch, PBAT, and potassium sorbate films. Finally, Rivero et al. (2013) demonstrated a positive effect on the conservation of pastry dough by applying chitosan film with propionic acid. As mentioned, most of the works used acids or salts as a preservative, and this reinforces the need for studies about films with essential oils in food preservation.

## Conclusion

In this work, it was possible to produce wheat flour/PBAT films incorporated with free and microencapsulated OEO by blow extrusion on a pilot scale, obtaining continuous and homogeneous films. The addition of free and microencapsulated OEO significantly increased the concentration of phenolic compounds and the antioxidant capacity of the films, being in greater proportion in the FOM film, confirming the protective effect of microencapsulation. The incorporation of encapsulated OEO provided less resistant films, with greater elongation and solubility in water. This fact was confirmed by the higher diffusion coefficient of OEO in 10% ethanol, which resulted in better control of molds and yeasts in fresh pastry during storage. Thus, the films produced in this work can be used as active biodegradable packaging when food contamination is predominantly caused by molds and yeasts, minimizing the use of synthetic additives in food and contributing to the reduction of solid waste in the environment.

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**Data Availability** Data and materials are available if necessary.

**Code Availability** Not applicable.

## Declarations

**Competing Interests** The authors declare no competing interests.

## References

Alvim, I. D., Stein, M. A., Koury, I. P., Dantas, F. B. H., & Cruz, C. L. C. V. (2016). Comparison between the spray drying and spray chilling microparticles contain ascorbic acid in a baked product application. *LWT - Food Science and Technology, 65*, 689–694. https://doi.org/10.1016/j.lwt.2015.08.049.

Andrade-Molina, T. P. d. C., Shirai, M. A., Grossmann, M. V. E., & Yamashita, F. (2013). Active biodegradable packaging for fresh pasta. *LWT - Food Science and Technology, 54*(1), 25–29. https://doi.org/10.1016/j.lwt.2013.05.011.

ASTM. (2000). Standard test methods for water vapor transmission of materials ASTM E96 - 00. In *Annual book of ASTM*. American Society for Testing and Materials.

ASTM. (2001). Standard test methods for tensile properties of thin plastic sheeting D882-00. In *In Annual book of ASTM*. American Society for Testing and Materials.

Benavides, S., Villalobos-Carvajal, R., & Reyes, J. E. (2012). Physical, mechanical and antibacterial properties of alginate film: Effect of the crosslinking degree and oregano essential oil concentration. *Journal of Food Engineering, 110*(2), 232–239. https://doi.org/10.1016/j.jfoodeng.2011.05.023.

Benincasa, P., Dominici, F., Bocci, L., Governatori, C., Panfili, I., Tosti, G., Torre, L., & Puglia, D. (2017). Relationships between wheat flour baking properties and tensile characteristics of derived thermoplastic films. *Industrial Crops and Products, 100*, 138–145. https://doi.org/10.1016/j.indcrop.2017.02.021.

Benzie, I. F. F., & Strain, J. J. (1996). The ferric reducing ability of plasma (FRAP) as a measure of "antioxidant power": The FRAP assay. *Analytical Biochemistry, 239*(1), 70–76. https://doi.org/10.1006/abio.1996.0292.

Bertuzzi, M. A., Castro Vidaurre, E. F., Armada, M., & Gottifredi, J. C. (2007). Water vapor permeability of edible starch based films. *Journal of Food Engineering, 80*(3), 972–978. https://doi.org/10.1016/j.jfoodeng.2006.07.016.

Boskabady, M. H., Tabatabaee, A., & Jalali, S. (2014). Potential effect of the extract of Zataria multiflora and its constituent, carvacrol, on lung pathology, total and differential WBC, IgE and eosinophil peroxidase levels in sensitized guinea pigs. *Journal of Functional Foods, 11*, 49–61. https://doi.org/10.1016/j.jff.2014.08.021.

Byun, Y., Kim, Y. T., & Whiteside, S. (2010). Characterization of an antioxidant polylactic acid (PLA) film prepared with α-tocopherol, BHT and polyethylene glycol using film cast extruder. *Journal of Food Engineering, 100*(2), 239–244. https://doi.org/10.1016/j.jfoodeng.2010.04.005.

Cardoso, L. G., Pereira Santos, J. C., Camilloto, G. P., Miranda, A. L., Druzian, J. I., & Guimarães, A. G. (2017). Development of active films poly (butylene adipate co-terephthalate) — PBAT incorporated with oregano essential oil and application in fish fillet preservation. *Industrial Crops and Products, 108*(September), 388–397. https://doi.org/10.1016/j.indcrop.2017.06.058.

da Costa, J. M. G., Borges, S. V., Hijo, A. A. C. T., Silva, E. K., Marques, G. R., Cirillo, M. Â., & de Azevedo, V. M. (2013). Matrix structure selection in the microparticles of essential oil oregano produced by spray dryer. *Journal of Microencapsulation, 30*(8), 717–727. https://doi.org/10.3109/02652048.2013.778909.

da Silva, T. B. V., Moreira, T. F. M., de Oliveira, A., Bilck, A. P., Gonçalves, O. H., Ferreira, I. C. F. R., et al. (2019). Araucaria angustifolia (Bertol.) Kuntze extract as a source of phenolic compounds in TPS/PBAT active films. *Food & Function, 10*(12), 7697–7706. https://doi.org/10.1039/C9FO01315F.

de Campos, S. S., de Oliveira, A., Moreira, T. F. M., da Silva, T. B. V., da Silva, M. V., Pinto, J. A., Bilck, A. P., Gonçalves, O. H., Fernandes, I. P., Barreiro, M. F., Yamashita, F., Valderrama, P., Shirai, M. A., & Leimann, F. V. (2019). TPCS/PBAT blown extruded films added with curcumin as a technological approach for active packaging materials. *Food Packaging and Shelf Life, 22*(November 2018), 100424. https://doi.org/10.1016/j.fpsl.2019.100424.

de Medeiros, J. A. S., Blick, A. P., Galindo, M. V., Alvim, I. D., Yamashita, F., Ueno, C. T., Shirai, M. A., Grosso, C. R. F., Corradini, E., & Sakanaka, L. S. (2019). Incorporation of oregano essential oil microcapsules in starch-poly (butylene adipate co-terephthalate) (PBAT) films. *Macromolecular Symposia, 383*(1), 1–7. https://doi.org/10.1002/masy.201800052.

de Souza, K. C., Correa, L. G., da Silva, T. B. V., Moreira, T. F. M., de Oliveira, A., Sakanaka, L. S., Dias, M. I., Barros, L., Ferreira, I. C. F. R., Valderrama, P., Leimann, F. V., & Shirai, M. A. (2020). Soy protein isolate films incorporated with Pinhão (Araucaria angustifolia (Bertol.) Kuntze) extract for potential use as edible oil active packaging. *Food and Bioprocess Technology, 13*(6), 998–1008. https://doi.org/10.1007/s11947-020-02454-5.

dos Santos Paglione, I., Galindo, M. V., de Medeiros, J. A. S., Yamashita, F., Alvim, I. D., Ferreira Grosso, C. R., Sakanaka, L. S., & Shirai, M. A. (2019). Comparative study of the properties of soy protein concentrate films containing free and encapsulated oregano essential oil. *Food Packaging and Shelf Life, 22*(October), 100419. https://doi.org/10.1016/j.fpsl.2019.100419.

Garcia, P. S., Grossmann, M. V. E., Shirai, M. A., Lazaretti, M. M., Yamashita, F., Muller, C. M. O., & Mali, S. (2014). Improving action of citric acid as compatibiliser in starch/polyesterblown films. *Industrial Crops and Products, 52*, 305–312. https://doi.org/10.1016/j.indcrop.2013.11.001.

Gontard, N., Duchez, C., Cuq, J.-L., & Guilbert, S. (1994). Edible composite films of wheat gluten and lipids: Water vapour permeability and other physical properties. *International Journal of Food Science & Technology, 29*(1), 39–50. https://doi.org/10.1111/j.1365-2621.1994.tb02045.x.

Ke, J., Xiao, L., Yu, G., Wu, H., Shen, G., & Zhang, Z. (2019). The study of diffusion kinetics of cinnamaldehyde from corn starch-based film into food simulant and physical properties of antibacterial polymer film. *International Journal of Biological Macromolecules, 125*, 642–650. https://doi.org/10.1016/j.ijbiomac.2018.12.094.

Leblanc, N., Saiah, R., Beucher, E., Gattin, R., Castandet, M., & Saiter, J.-M. (2008). Structural investigation and thermal stability of new extruded wheat flour based polymeric materials. *Carbohydrate Polymers, 73*(4), 548–557. https://doi.org/10.1016/j.carbpol.2007.12.034.

Lee, J.-H., Yang, H.-J., Lee, K.-Y., & Song, K. B. (2016). Physical properties and application of a red pepper seed meal protein composite film containing oregano oil. *Food Hydrocolloids, 55*, 136–143. https://doi.org/10.1016/j.foodhyd.2015.11.013.

Li, B., Kennedy, J. F., Peng, J. L., Yie, X., & Xie, B. J. (2006). Preparation and performance evaluation of glucomannan–chitosan–nisin ternary antimicrobial blend film. *Carbohydrate Polymers, 65*(4), 488–494. https://doi.org/10.1016/j.carbpol.2006.02.006.

Liu, Q. R., Wang, W., Qi, J., Huang, Q., & Xiao, J. (2019). Oregano essential oil loaded soybean polysaccharide films: Effect of Pickering type immobilization on physical and antimicrobial properties. *Food Hydrocolloids, 87*(June 2018), 165–172. https://doi.org/10.1016/j.foodhyd.2018.08.011.

Lv, J., Yu, L., Lu, Y., Niu, Y., Liu, L., Costa, J., & Yu, L. (2012). Phytochemical compositions, and antioxidant properties, and antiproliferative activities of wheat flour. *Food Chemistry, 135*(2), 325–331. https://doi.org/10.1016/j.foodchem.2012.04.141.

Mensor, L. L., Menezes, F. S., Leitão, G. G., Reis, A. S., dos Santos, T. C., Coube, C. S., & Leitão, S. G. (2001). Screening of Brazilian plant extracts for antioxidant activity by the use of DPPH free radical method. *Phytotherapy Research, 15*(2), 127–130. https://doi.org/10.1002/ptr.687.

Mücke, N., da Silva, T. B. V., de Oliveira, A., Moreira, T. F. M., Venancio, C. D. S., Marques, L. L. M., Valderrama, P., Gonçalves, O. H., da Silva-Buzanello, R. A., Yamashita, F., Shirai, M. A., Genena, A. K., & Leimann, F. V. (2021). Use of water-soluble curcumin in TPS/PBAT packaging material: Interference on reactive extrusion and oxidative stability of chia oil. *Food and Bioprocess Technology, 14*(3), 471–482. https://doi.org/10.1007/s11947-021-02584-4.

Mushtaq, M., Gani, A., Gani, A., Punoo, H. A., & Masoodi, F. A. (2018). Use of pomegranate peel extract incorporated zein film with improved properties for prolonged shelf life of fresh Himalayan cheese (Kalari/kradi). *Innovative Food Science & Emerging Technologies, 48*, 25–32. https://doi.org/10.1016/j.ifset.2018.04.020.

Neltner, T. G., Alger, H. M., Leonard, J. E., & Maffini, M. V. (2013). Data gaps in toxicity testing of chemicals allowed in food in the United States. *Reproductive Toxicology, 42*, 85–94. https://doi.org/10.1016/j.reprotox.2013.07.023.

Ouattara, B., Simard, R. E., Piette, G., Bégin, A., & Holley, R. A. (2000). Inhibition of surface spoilage bacteria in processed meats by application of antimicrobial films prepared with chitosan. *International Journal of Food Microbiology, 62*(1), 139–148. https://doi.org/10.1016/S0168-1605(00)00407-4.

Pelissari, F. M., Grossmann, M. V. E., Yamashita, F., & Pineda, E. A. G. (2009). Antimicrobial, mechanical, and barrier properties of cassava starch−chitosan films incorporated with oregano essential oil. *Journal of Agricultural and Food Chemistry, 57*(16), 7499–7504. https://doi.org/10.1021/jf9002363.

Puglia, D., Dominici, F., Kenny, J. M., Santulli, C., Governatori, C., Tosti, G., & Benincasa, P. (2016). Tensile behavior of thermoplastic films from wheat flours as function of raw material baking properties. *Journal of Polymers and the Environment, 24*(1), 37–47. https://doi.org/10.1007/s10924-015-0745-4.

Ribeiro-Santos, R., Andrade, M., & Sanches-Silva, A. (2017). Application of encapsulated essential oils as antimicrobial agents in food packaging. *Current Opinion in Food Science, 14*, 78–84. https://doi.org/10.1016/j.cofs.2017.01.012.

Ribeiro-Santos, R., Andrade, M., Sanches-Silva, A., & de Melo, N. R. (2018). Essential oils for food application: Natural substances with established biological activities. *Food and Bioprocess Technology, 11*(1), 43–71. https://doi.org/10.1007/s11947-017-1948-6.

Rivero, S., Giannuzzi, L., García, M. A., & Pinotti, A. (2013). Controlled delivery of propionic acid from chitosan films for pastry dough conservation. *Journal of Food Engineering, 116*(2), 524–531. https://doi.org/10.1016/j.jfoodeng.2012.12.025.

Ruiz-Navajas, Y., Viuda-Martos, M., Sendra, E., Perez-Alvarez, J. A., & Fernández-López, J. (2013). In vitro antibacterial and antioxidant properties of chitosan edible films incorporated with Thymus moroderi or Thymus piperella essential oils. *Food Control, 30*(2), 386–392. https://doi.org/10.1016/j.foodcont.2012.07.052.

Selzer, D., Abdel-Mottaleb, M. M. A., Hahn, T., Schaefer, U. F., & Neumann, D. (2013). Finite and infinite dosing: Difficulties in measurements, evaluations and predictions. *Advanced Drug Delivery Reviews, 65*(2), 278–294. https://doi.org/10.1016/j.addr.2012.06.010.

Silveira, M. F. A., Soares, N. F. F., Geraldine, R. M., Andrade, N. J., Botrel, D. A., & Gonçalves, M. P. J. (2007). Active film incorporated with sorbic acid on pastry dough conservation. *Food Control, 18*(9), 1063–1067. https://doi.org/10.1016/j.foodcont.2006.07.004.

Singleton, V. L., & Rossi, J. A. (1965). Colorimetry of total phenolics with phosphomolybdic-phosphotungstic acid reagents. *American Journal of Enology and Viticulture, 16*(3), 144 LP–144158 http://www.ajevonline.org/content/16/3/144.abstract. Accessed 3 July 2020.

Solano, A. C. V., & de Gante, C. R. (2012). Two different processes to obtain antimicrobial packaging containing natural oils. *Food and Bioprocess Technology, 5*(6), 2522–2528. https://doi.org/10.1007/s11947-011-0626-3.

Sousa, G. M., Yamashita, F., & Soares Júnior, M. S. (2016). Application of biodegradable films made from rice flour, poly(butylene adipate-co-terphthalate), glycerol and potassium sorbate in the preservation of fresh food pastas. *LWT - Food Science and Technology, 65*, 39–45. https://doi.org/10.1016/j.lwt.2015.07.054.

Teodoro, R. A. R., de Barros Fernandes, R. V., Botrel, D. A., Borges, S. V., & de Souza, A. U. (2014). Characterization of microencapsulated rosemary essential oil and its antimicrobial effect on fresh dough.

*Food and Bioprocess Technology, 7*(9), 2560–2569. https://doi.org/10.1007/s11947-014-1302-1.

Thaipong, K., Boonprakob, U., Crosby, K., Cisneros-Zevallos, L., & Hawkins Byrne, D. (2006). Comparison of ABTS, DPPH, FRAP, and ORAC assays for estimating antioxidant activity from guava fruit extracts. *Journal of Food Composition and Analysis, 19*(6–7), 669–675. https://doi.org/10.1016/j.jfca.2006.01.003.

Toledo Hijo, A. A. C., Da Costa, J. M. G., Silva, E. K., Azevedo, V. M., Yoshida, M. I., & Borges, S. V. (2015). Physical and thermal properties of oregano (*Origanum vulgare* L.) essential oil microparticles. *Journal of Food Process Engineering, 38*(1), 1–10. https://doi.org/10.1111/jfpe.12120.

Viuda-Martos, M., El Gendy, A. E.-N. G. S., Sendra, E., Fernández-López, J., Abd El Razik, K. A., Omer, E. A., & Pérez-Alvarez, J. A. (2010). Chemical composition and antioxidant and anti-Listeria activities of essential oils obtained from some Egyptian plants. *Journal of Agricultural and Food Chemistry, 58*(16), 9063–9070. https://doi.org/10.1021/jf101620c.

Zehetmeyer, G., Meira, S. M. M., Scheibel, J. M., de Oliveira, R. V. B., Brandelli, A., & Soares, R. M. D. (2016). Influence of melt processing on biodegradable nisin-PBAT films intended for active food packaging applications. *Journal of Applied Polymer Science, 43212*(13), 1–10. https://doi.org/10.1002/app.43212.

Zhai, X., Wang, W., Zhang, H., Dai, Y., Dong, H., & Hou, H. (2020). Effects of high starch content on the physicochemical properties of starch/PBAT nanocomposite films prepared by extrusion blowing. *Carbohydrate Polymers, 239*, 116231. https://doi.org/10.1016/j.carbpol.2020.116231.