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# HYDROALCOHOLIC EXTRACTION, ULTRASOUND-ASSISTED EXTRACTION AND ENCAPSULATION OF BIOACTIVES FROM VEGETABLE RESIDUES

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Dissertação de Mestrado do Programa de Pós-Graduação em Alimentos e Nutrição da Universidade Federal do Estado do Rio de Janeiro, como requisito parcial para obtenção do título de Mestre em Ciência dos Alimentos.

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# RESUMO

Compostos bioativos são biomoléculas que possuem ação potencial na manutenção da saúde e prevenção de doenças. Assim, há um crescente interesse na extração e aplicação destes compostos. Diferentes técnicas podem ser aplicadas na extração de compostos bioativos a partir de diversas fontes vegetais, estando entre elas as extrações com solventes orgânicos e as extrações assistidas por ultrassom. Há ainda um crescente interesse pela exploração e reutilização de resíduos vegetais como fonte de compostos bioativos, tendo em vista sua vasta formação durante as etapas de produção de alimentos. Entretanto, compostos bioativos são sensíveis às condições ambientais, e sugere-se algum meio de preservação destes compostos. Dentre esses meios, está a secagem por aspersão ou *spray drying*. Desta forma, os objetivos dessa desta Dissertação são apresentar os principais materiais de parede utilizados na encapsulação de compostos bioativos na secagem por aspersão e realizar a extração por etapas de compostos bioativos a partir de uma farinha obtida de resíduo vegetal, utilizando extração hidroalcoólica e extração assistida por ultrassom. Esta dissertação será apresentada na forma de dois artigos científicos. O primeiro, discute as características físico-químicas dos principais materiais de parede utilizados para a secagem de bioativos via *spray drying*. O segundo, discute as interações fisicoquímicas e correlações entre a extração de bioativos pelos processos de extração hidroalcoólica e assistida por ultrassom e as modificações microestruturais que ocorrem na matriz vegetal, culminando na sua posterior encapsulação, conforme segue: O teor total de polifenóis na Farinha de Frutas e Hortaliças (FVF) foi de 76.28 ± 2.17 a 92.32 ± 5.79 mg equivalentes de ácido gálico por grama de FVF para a extração hidroalcoólica, e de 113.02 ± 2.71 a 134.48 ± 1.66 mg equivalentes de ácido gálico por grama de FVF para a extração assistida por ultrassom. A microestrutura da FVF sofreu mudanças significativas após o tratamento em cada tipo de extração e condição aplicada. A extração assistida por ultrassom foi o tipo que mais promoveu alterações na microestrutura da FVF. Por fim, o teor de polifenóis nas cápsulas produzidas variou entre 0.54 ± 0.04 e 1.92 ± 0.04 mg $GAEg^{-1}$ equivalentes de ácido gálico por grama de pó. As cápsulas possuíam forma esférica e acidentada, porém sem rachaduras ou quebras, indicando um bom processo de encapsulação.



**Palavras-chave:** Extração Hidroalcoólica. Extração com ultrassom. *Spray drying*.

# ABSTRACT

Bioactive compounds are biomolecules that have a potential action in maintaining health and preventing diseases and there is a growing interest in the extraction and application of these compounds. Different techniques can be applied in the extraction of bioactive compounds from different plant sources, including extractions with organic solvents and ultrasound-assisted extraction. There is also a growing interest in the exploration and reuse of vegetable waste as a source of bioactive compounds, in view of its vast formation during the stages of food production. However, bioactive compounds are sensitive to environmental conditions, and some means of preserving these compounds is suggested. Among these means is *spray drying*. Thus, the objectives of this study are to present the main wall materials used in the encapsulation of bioactive compounds by *spray drying* and to perform a multi-step extraction of bioactive compounds from a flour produced from vegetable waste, using hydroalcoholic extraction and ultrasound-assisted extraction. It will be presented in the form of two articles. The first discusses the physicochemical characteristics of the main wall materials used for drying bioactive materials via *spray drying*. The second discusses the physicochemical interactions and correlations between bioactive extraction by hydroalcoholic and ultrasound-assisted extraction processes and the microstructural changes that occur in the plant matrix, culminating in their subsequent encapsulation, as follows: The total polyphenol content of the extracts were 76.28 ± 2.17 to 92.32 ± 5.79 mg of gallic acid equivalent.$g^{-1}$ FVF for HAE and 113.02 ± 2.71 to 134.48 ± 1.66 mg of gallic acid equivalent.$g^{-1}$ FVF. The microstructure of the FVF had significantly changes post treatment on each extraction type. The UAE was the extraction type that most modified the microstructure of the FVF, increasing the release of bound polyphenols. The capsules had TPC varied from 0.54 ± 0.04 to 1.92 ± 0.04 mg of gallic acid equivalent.$g^{-1}$ of powder. They had spherical and tipped morphology with no cracks, suggesting a good encapsulation process.



**Keywords:** Hydroalcoholic extraction. Ultrasound-assisted extraction. *Spray drying*.

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# SUMÁRIO

# 1 INTRODUÇÃO

Resíduos de alimentos são alimentos inteiros ou partes que foram descartados ou perdidos e que não foram reciclados ou utilizados para outros fins. Sua formação pode ocorrer em todas as etapas da cadeia de produção e é considerada um problema global da atualidade pois o acúmulo desta matéria orgânica promove grande impacto ambiental custando bilhões de dólares por ano para seu manejo (Girotto, Alibardi, & Cossu, 2015; Lin et al., 2013).

Atualmente, encontra-se vigente no Brasil o Plano Nacional de Resíduos Sólidos, previsto na Lei nº 12.305/2010, que institui a Política Nacional de Resíduos Sólidos. Dentre as vertentes do plano, o reaproveitamento de resíduos de alimentos gerados em processos da agricultura e agroindústria vem sendo incentivado nacionalmente para o uso como biomassa para produção de adubo e biocombustíveis (IPEA, 2012).

Entretanto, uma vez que os resíduos de alimentos são, em sua maioria, partes vegetais usualmente não consumidas (como talos, folhas e cascas) tão ricos em nutrientes (como vitaminas, minerais e outros compostos bioativos) quanto as partes usualmente consumidas, sua utilização em processos tecnológicos para a indústria de alimentos vem sendo incentivada por contribuir para a redução dos custos de manejo e o impacto ambiental gerado pelo acúmulo desses resíduos (Lin et al., 2013; A. Nesterenko, Alric, Silvestre, & Durrieu, 2014; Taghvaei & Jafari, 2015).

A extração de compostos bioativos de resíduos vegetais vai ao encontro de políticas públicas vigentes no país, auxiliando na redução do impacto ambiental gerado por estes resíduos, bem como fornecendo matérias-primas de baixo custo para a obtenção de novos produtos com potencial benefício à saúde (IPEA, 2012).

Após a extração, uma vez que os compostos antioxidantes são sensíveis a condições ambientais diversas, inúmeros tipos de processamentos tecnológicos têm sido pesquisados para a conservação destes compostos visando o aumento do tempo de prateleira, por exemplo, a encapsulação (Pinho, Grootveld, Soares, & Henriques, 2014).

O processo de encapsulação é definido como um método de proteção para compostos ativos que são sensíveis a condições ambientais de forma que estes compostos

sejam envolvidos em uma membrana, denominada encapsulante, cobertura ou parede, resistente a essas condições, dando uma sobrevida aos compostos encapsulados e permitindo que seu conteúdo seja liberado apenas no momento desejado (Moser et al., 2017; Quirós-Sauceda, Ayala-Zavala, Olivas, & González-Aguilar, 2014).

Atualmente, o método mais comum e de menor custo para realização de encapsulação de aditivos alimentares se dá através da técnica de secagem por aspersão ou *spray drying*. Este método consiste em remover a fração líquida de uma amostra que se encontra em estado fluido, seja ela uma solução, emulsão, suspensão ou pasta, adicionando um material encapsulante, através do uso de ar quente, formando microcápsulas em pó, promovendo a concentração da amostra (Keshani, Daud, Nourouzi, Namvar, & Ghasemi, 2015).

A escolha do material encapsulante deve levar em consideração uma série de fatores, como: não ser tóxico; ser capaz de passar por processos de esterilização; possuir estabilidade físico-química; deve fornecer resistência mecânica ao material encapsulado; ser resistente a luz, oxigênio, variações de temperatura, umidade dentre outras condições ambientais (Gharsallaoui, Roudaut, Chambin, Voilley, & Saurel, 2007; Tolve et al., 2016).

A maior parte dos compostos bioativos são moléculas produzidas pelo metabolismo secundário das células vegetais e podem se apresentar como polifenóis livres ou polifenóis conjugados (S. Liu et al., 2019; Pinaffi et al., 2020), com o segundo tipo formando complexos com polissacarídeos amiláceos e não amiláceos, como a celulose, hemicelulose, pectina (F. Zhu, 2018) e proteínas (S. Liu et al., 2019). Os polifenóis se ligam a estrutura dos polissacarídeos não-amiláceos frequentemente por pontes de hidrogênio através de seus grupamentos hidroxila e por interações por hidrofobicidade (D. Liu, Lopez-Sanchez, Martinez-Sanz, Gilbert, & Gidley, 2019), na superfície e no interior de polímeros vegetais, em sítios específicos presentes em sua microestrutura (Jakobek & Matić, 2019; F. Zhu, 2018).

Assim, para extrair os polifenóis de matrizes vegetais, diferentes tipos de extração sólido-líquido são realizados, como as extrações convencionais com solventes orgânicos (Pinaffi et al., 2020; F. Zhu, 2018) ou técnicas consideradas tecnologicamente mais

recentes, como a extração assistida por ultrassom (Balasubramaniam, Ayyappan, Sathvika, & Antony, 2019; Maran, Manikandan, Nivetha, & Dinesh, 2017).

Devido as interações hidrofóbicas dos polifenóis com a estrutura polimérica dos vegetais, o uso de solventes orgânicos na extração convencional objetiva reduzir a afinidade dos polifenóis com a estrutura polimérica, permitindo seu carreamento e, para extrair compostos com diferentes polaridades, o solvente orgânico pode ser homogeneizado com outro solvente orgânico com polaridade diferente ou água (Acosta-Estrada, Gutiérrez-Uribe, & Serna-Saldívar, 2014; F. Zhu, 2018). Assim, a composição polimérica da matrix vegetal torna-se um parâmetro relevante a ser observado durante a extração de polifenóis conjugados, uma vez que eles interagem de diferentes maneiras com a matrix (Jakobek & Matić, 2019).

Desta forma, os objetivos dessa desta Dissertação são apresentar os principais materiais de parede utilizados na encapsulação de compostos bioativos por *spray drying* e realizar a extração por etapas de compostos bioativos a partir de uma farinha produzida a partir de resíduo vegetal, utilizando extração hidroalcoólica e extração assistida por ultrassom. Esta dissertação será apresentada na forma de dois artigos, inicialmente uma revisão bibliográfica relacionada a materiais de parede aplicados à encapsulação de compostos bioativos por *spray drying* e o segundo artigo avaliará a extração continua de bioativos de uma matriz vegetal através de extração hidroalcoólica e extração assistida por ultrassom observando a microestrutura vegetal, culminando na encapsulação dos bioativos.

# 2 *SPRAY DRYING* WALL MATERIALS: RELATIONSHIP WITH BIOACTIVE COMPOUNDS

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**- REVIEW ARTICLE -**

**Abstract**

The encapsulation by *spray drying* is a common method of encapsulation on the food industry. Many biopolymers are described on the literature as wall materials for this purpose. As different wall materials are applied on *spray drying*, it is necessary to know their physicochemical characteristics. In view of the different applications for this technology, the objective of the present study is to discuss the physicochemical characteristics of several wall materials used in encapsulation of bioactive compounds by *spray drying* for human consumption. Among all biopolymers make blends of carbohydrate-based and protein-based biopolymers seems to be the better way to encapsulate bioactive compounds, especially when using vegetable proteins in view of reach a less allergenic product.

**Keywords**: biopolymer; *spray drying*; bioactive compounds; wall materials

## 2.1 Introduction

The encapsulation by the *spray drying* is the most common method of encapsulation on the food industry (Keshani et al., 2015). On *spray drying*, compounds that are sensitive to environmental conditions can be encapsulated with aid of a wall material, giving to the physicochemical properties from the selected wall material a high importance on the process (Jafari, Assadpoor, He, & Bhandari, 2008). The ability to form polymers, to resist mechanical stress and environmental conditions (e.g. air moisture, temperature, and water activity) are some of these vital characteristics (Tolve et al., 2016). It also must be non-toxic and non-allergenic for humans, able to be sterilized and have no or a mild taste, which will allow its use for encapsulation of food ingredients (Fernandes, Borges, & Botrel, 2014; Jafari et al., 2008; Shankar, Wang, & Rhim, 2017; Tolve et al., 2016).

The wall material can range from high molecular weight biopolymers, like starch, modified starch and proteins, to lower molecular weight biopolymers as cyclodextrins and maltodextrins (Pinho et al., 2014; Samantha et al., 2015).

On the other hand, there's a growing interest on the application of this technology on compounds like polyphenols, carotenoids, flavonoids, and anthocyanins as these molecules have a high biological activity, including antioxidant capacities, that increases the interest of its use on food products (Ferreira et al., 2013; Jafari et al., 2008; Lu, Kelly, & Miao, 2016; Pereira, 2014). For this purpose, the mostly used biopolymers are maltodextrin, gum arabic and milk proteins (Samantha et al., 2015; Shamaei, Seiiedlou, Aghbashlo, Tsotsas, & Kharaghani, 2017).

Naturally, interactions may occur, in order to stabilize the microcapsules and to protect the core. The interactions can be: 1) between the polymer and the bioactive, where the wall material enhance the stability of the core, preventing loss and/or formation of less stable forms; 2) a polymer-polymer interaction, where the biopolymer is previously modified to reach better efficiency on the process; 3) synergistic interactions between two or more polymers, where blend of biopolymers is applied on the process to reach better physicochemical properties for the microcapsule (Fang & Bhandari, 2010; Labuschagne, 2018).

In view of the different applications for this technology, the objective of the present study is to discuss the physicochemical characteristics of several wall materials used in encapsulation of bioactive compounds by *spray drying* for human consumption.

## 2.2 Polysaccharide-based wall materials

As mentioned before, several carbohydrate-based biopolymers are used as wall material on bioactives encapsulation. On this process, the glycosidic bonds give the carbohydrate polymer unique physicochemical characteristics, stability, and resistance of environmental conditions, according to their angular (with α-(1-4) glycosidic bond) or linear (with β-(1-4) glycosidic bond) tri-dimensional configuration (Gray et al., 2017; C. Zhu, Krumm, Facas, Neurock, & Dauenhauer, 2017). The main carbohydrates used as wall materials and their relationship with the core are discussed below.

### *2.2.1 Starch, modified starch and its derivates*

The starch has the advantage of being a low-cost material with good hydrophilic content retention capabilities (F. Zhu, 2017). It has a low viscosity at high solids concentration and a mild flavor and its retention capacity is directly linked to the proportion of dextrose in the hydrolysed starch, since the permeability of the oxygen in the membrane will be inversely proportional to the concentration of dextrose in the same, resulting in a greater efficiency of retention and protection of the encapsulated content (Gupta, Chawla, Arora, Tomar, & Singh, 2015; Jafari et al., 2008). The ability of the starch granules may be involved in the protection of the bioactive encapsulated since the granule act as a physical barrier that entraps the bioactive, reducing its exposition to the environment (F. Zhu, 2017).

#### *2.2.1.1 Modified starch*

The starch does not have good emulsifying capabilities, which results in a low retention capacity of volatile compounds and other hydrophobic character bioactives during spray drying. The literature indicates that starch esterified with n-octenyl succinyl anhydride (OSA) has high retention capacity of these molecules and a low number of compounds adhered to the external surface, due to the significant improvement that the esterification gives to its emulsifying capacities (Agama-Acevedo & Bello-Perez, 2017).

This improvement would be provided due to the presence of hydrophobic clusters given by the octenyl group obtained by esterification, and by the presence of hydrophilic groups such as carboxylates or sodium carboxylates, originating from starch (Abbas, K. Khalil, & Meor Hussin, 2010; Sarkar & Singhal, 2011; Soottitantawat, Partanen, Neoh, & Yoshii, 2015).

Although, the esterification of starch is able to change rheological characteristics of the molecule (Sweedman, Tizzotti, Schäfer, & Gilbert, 2013). The substitution of hydroxyl groups by the OSA disrupts the crystalline structure of the starch granule by weakening the hydrogen bonds, reflecting on a reduce of the gelatinization enthalpy (Altuna, Herrera, & Foresti, 2018) and a reduction of pasting temperature (Bello-Flores, Nuñez-Santiago, Martín-Gonzalez, BeMiller, & Bello-Pérez, 2014). Since the esterification can occur on carbons 2, 3 or 6 of the glucose molecule, this process with OSA can also reduce the digestibility of the starch by a reduction of interaction between amylases and the starch (Zhang, Mei, Chen, & Chen, 2017; Zheng et al., 2017). Thus it reaches the colon, being fermented by the microbiota, which benefices the colon health (Zhang et al., 2017).

*2.2.1.2 Maltodextrin*

Maltodextrins are products of starch hydrolysis, consisting of α-D-glucose units linked mainly by α-(1-4) glycosidic bonds and described by their dextrose equivalency (DE) since the DE is inversely related to their average molecular weight (Labuschagne, 2018). It is used on the food industry for various purposes and have multiple benefits for food processing like: high solubility in water, low viscosity, bland flavour, is colourless when in solution, offers protection to oxidation when used as wall material, and have a low cost of obtention (Saavedra-Leos, Leyva-Porras, Araujo-Díaz, Toxqui-Terán, & Borrás-Enríquez, 2015; Tonon et al., 2009).

It does not present a good emulsifying property due to the presence of hydroxyl groups that favor wettability, creating a limiting feature on encapsulation (Waterhouse, Sun-Waterhouse, Su, Zhao, & Zhao, 2017a). This can be changed with the addition of other wall materials, like pectin (Sansone et al., 2011), gum arabic (Fernandes et al., 2014), whey protein (Moser et al., 2017), or by esterification (Agama-Acevedo & Bello-

Perez, 2017). These changes will contribute with emulsification by reducing the presence of hydroxyl groups (as esterification) or by forming complexes that allow the added wall to act as an emulsion stabilizer (Faridi Esfanjani, Jafari, & Assadpour, 2017).

Maltodextrin can also be used in combination with other encapsulating materials to reduce the loss of volatile compounds during the drying process (Soottitantawat et al., 2015) even on higher inlet temperatures (about 150ºC) since it creates a wall on the drop surface, improving the powder yield and lower moisture on final product (Paini et al., 2015; Santhalakshmy, Don Bosco, Francis, & Sabeena, 2015). It can also support great variations on water activity being able to maintain their polymeric structure until the water activity of 0,695 was reached (Araujo-Díaz, Leyva-Porras, Aguirre-Bañuelos, Álvarez-Salas, & Saavedra-Leos, 2017). This resistance to water activity variations after the encapsulation process may be due to the smaller size of the molecule and the lesser presence of hydroxyl groups, which results on a lower solubility and adsorption of air humidity (Tonon et al., 2009; Zotarelli, da Silva, Durigon, Hubinger, & Laurindo, 2017). In addition, it can promote an additional stabilizing effect due to its ability to reduce the bioactive mobility and reducing the permeability of the oxygen inversely proportional to its DE (Gupta et al., 2015). However, it is not able to prevent the microcapsules from wilting during the drying process, providing the microcapsules with a somewhat spherical and tipped morphology (Saénz, Tapia, Chávez, & Robert, 2009).

*2.2.1.3 Cyclodextrins (CD)*

Cyclodextrins are oligosaccharides composed of 6-8 glycosides obtained from the degradation of the starch, forming rigid and circular structures with a highly hydrophilic exterior and a low hydrophilic site inside the molecule (Pinho et al., 2014). Studies have shown that walls composed of these oligosaccharides are more resistant to oxidation and pH variations and are therefore well researched on the encapsulation of essential oils and extracts of bioactive compounds of vegetable origin (Kalogeropoulos, Yannakopoulou, Gioxari, Chiou, & Makris, 2010; Pasrija, Ezhilarasi, Indrani, & Anandharamakrishnan, 2015).

Due to their molecular structure, cyclodextrins are promising materials for the encapsulation of hydrophobic bioactive compounds and flavors, since it is able to retain

these molecules in their low polar interior by hydrophobic forces or van der Waals interactions, protecting them from oxidation and other external environmental conditions and, in addition, upgrading their water solubility (Carlotti, Sapino, Ugazio, & Caron, 2011; Pinho et al., 2014).

Actually, CDs have some limitations for their use. The regulatory status of the CDs differs between countries but only the β-CD is approved for use worldwide as a wall material (carrier agent) by FAO/WHO (FAO/WHO, 2019). On the other hand, there are not many applications nowadays for the food industry since they're less soluble in water then the linear biopolymers as starch and maltodextrin (Astray, Gonzalez-Barreiro, Mejuto, Rial-Otero, & Simal-Gándara, 2009). Although, the β-CD may be used for encapsulation of bioactive compounds aiming the application on apolar medium because of its low water solubility.

*2.2.2 Gum Arabic*

Gum arabic is a biopolymer consisting of d-glucuronic acid, l-rhamnose, d-galactose and l-arabinose, with approximately 2% of associate protein content. Its high solubility on water, low viscosity, and high glass transition temperature are characteristics that guided this molecule to be the most common wall material used for encapsulation purposes. Unfortunately, it has a high cost and limited availability (Comunian et al., 2011; Gupta et al., 2015; Tonon et al., 2009).

Microcapsules made of gum arabic by spray drying are stable at low water activities and maintain the stability of core on a 200 days shelf-life (Ramírez, Giraldo, & Orrego, 2015). However, in water activities higher then 0,74 it loses the stability and starts a dissolution process of the wall, leading to core loss (Rascón, Beristain, García, & Salgado, 2011).

The protein portion on gum arabic structure may act as stabilizers and emulsifiers, which provides the good ability to create microcapsules and is related to emulsify with all main oils (Ramírez et al., 2015; Samantha et al., 2015). It's structure is able to promote aggregation with bioactives, probably retaining them on vacuoles-like structures (Dordevic et al., 2014). Unfortunately, the high ramified structure of gum arabic may act

as a semi-permeable membrane for the oxygen, leading to a partial protection against oxidation (Samantha et al., 2015).

### *2.2.3 Inulin*

Inulin is a natural linear polysaccharide made of β-(2-1) linked D-fructose units, that constitutes chains of different lengths and polymerization degrees that range from 10 to 60, with a glucose unit on the final portion of each chain (Bakowska-Barczak & Kolodziejczyk, 2011). It is more flexible than other carbohydrate polymers because of its chain structure where only one atom of the polymer attaches to the fructose ring (Barclay, Ginic-markovic, Cooper, & Petrovsky, 2010; Lacerda et al., 2016).

It has a notorious hydrophilic characteristic (Mensink, Frijlink, Van Der Voort Maarschalk, & Hinrichs, 2015), however, it was demonstrated that it is able to encapsulate hydrophobic molecules like anthocyanins (Lacerda et al., 2016), rosemary essential oil (Fernandes et al., 2014), black currant polyphenols (Bakowska-Barczak & Kolodziejczyk, 2011), and antioxidant-rich blueberry waste extracts (Waterhouse et al., 2017a). Although, inulin microcapsules are sensitive to environmental conditions such as temperature and air humidity, which leads to losing the core by agglomeration promoted by water adsorption (Daza et al., 2016). When used along another wall material, the difference on their glass transition temperatures can make inulin dry faster than the other wall material, creating a wall that can entrap the water inside the microcapsule (Castel, Rubiolo, & Carrara, 2018). On the other hand, although the faster drying process creates microcapsules with higher moisture, there is no influence of the hygroscopicity of the microcapsules, which is desirable for a longer shelf life (Botrel, de Barros Fernandes, Borges, & Yoshida, 2014).

### *2.2.4 Pectin*

Pectin is a polysaccharide from the plant cell walls, located on middle lamella, with a heterogeneous structure where their main structure is composed of galacturonic acid linked with α-(1-4) glycosidic bonds and can have several side chains containing sugars, like xylose, arabinose, glucose, mannose, and others (Müller-Maatsch et al., 2016; Sansone et al., 2011).

It was already reported that it can be used as an adjuvant on the encapsulation process with whey protein concentrate (Assadpour, Jafari, & Maghsoudlou, 2017; Mohammadi, Jafari, Esfanjani, & Akhavan, 2016), β-lactoglobulin (Serfert et al., 2013), maltodextrin (Sansone et al., 2011) and was able to preserve the bioactivity of the core material. Although, the metoxilation degree may play a role on the microcapsule formation (Sarkar & Singhal, 2011). The lesser presence of hydrophobic clusters like the methoxyl groups allow the low methoxylated pectin to interact by electrostatic and/or ionic bonds (Chan, Choo, Young, & Loh, 2017) with other biopolymers, like proteins (Tamm, Härter, Brodkorb, & Drusch, 2016) which may graduate the water loss during the spray drying process. As a side effect, the association of the pectin with proteins by electrostatic interactions creates a physical barrier that is able to reduce the oxidation induced by metal-lipid interactions of hydrophobic core materials, since it is able to chelate the metallic ions (Serfert et al., 2013; Tamm et al., 2016).

*2.2.5 Chitin and Chitosan*

Chitin is a natural biopolymer composed by N-acetyl-D-glucosamine (GluNAc) subunits liked by β-1,4 glycosidic bonds, obtained crustacean's exoskeletons (eg: crabs, shrimps, lobsters, and others), which is usually treated as food waste (Muxika, Etxabide, Uranga, Guerrero, & de la Caba, 2017). Chitin can be deacetylated creating a linear heteropolysaccharide with a cationic charge named as chitosan. This process doesn't change the type of linkage or the subunits the biopolymer have (Varun et al., 2017). They're biopolymers with high molecular weight and a notorious hydrophobic characteristic and has been used for encapsulation of both hydrophilic and hydrophobic molecules on food and drug industry, aiming a controlled release of the content (Aranaz, Paños, Peniche, Heras, & Acosta, 2017; Varun et al., 2017).

On encapsulation, chitosan was already used for encapsulate cobalamin and ascorbic acid, both hydrophilic vitamins, with a high yield (55% and 45%, respectively) even on low temperatures (120ºC on inlet and 65°C on outlet) and preserving the antioxidant effect of the encapsulated vitamins (Estevinho, Carlan, Blaga, & Rocha, 2016). It was also used to encapsulate glutathione to be incorporated as an antioxidant on wine (Webber, de Siqueira Ferreira, Barreto, Weiss-Angeli, & Vanderlinde, 2018) and encapsulate tea polyphenols aiming to enhance their bioavailability on the gastrointestinal

tract by an improvement of their stability and prevention of their oxidation (Liang et al., 2017).

*2.2.6 Other carbohydrate-based wall materials*

As the literature moves forward, new carbohydrates are related as wall materials on spray drying. Pectin of unconventional origins, cellulose, hydroxypropyl methylcellulose phthalate, polydextrose and new types of gums are some examples (Kuck & Noreña, 2016; Alla Nesterenko, Alric, Silvestre, & Durrieu, 2013; Tontul, Topuz, Ozkan, & Karacan, 2016; J. Wang, Li, Chen, Liu, & Chen, 2016) with some of them being related with superior encapsulation properties when compared with gum arabic, like mesquite gum (Rodea-González et al., 2012), while others can be obtained from food wastes, like pectin (Marić et al., 2018; Petkowicz, Vriesmann, & Williams, 2017). As new wall materials, more studies are needed to elucidate their properties and applications on spray drying.

**2.3 Protein-based wall materials**

The chemical properties from the proteins play a role in the encapsulation process. They can create films and polymers being used alone or in association with other molecules (mainly carbohydrates) acting as a physical wall and providing the retention of the core (Moser et al., 2017; A. Nesterenko et al., 2014; Sansone et al., 2011; Soottitantawat et al., 2015), or it can interact with the core by polar interactions (W. Liu, Chen, Cheng, & Selomulya, 2016; Tapal & Tiku, 2012).

It is worth mentioning that hydrophobic interactions occur due to the charge that the proteins and the core materials may have. The pH of the core material must be observed to determine the pH of the protein solution that will be used to encapsulate it, since at pH below the isoelectric point the protein assumes positive charge and above the isoelectric point, it assumes a negative charge (Lam & Nickerson, 2013; Wihodo & Moraru, 2013). This must be observed when creating wall materials blends because the charge change may create conditions for the carbohydrates interact with the proteins by non-covalent interactions (Rodriguez Patino & Pilosof, 2011). When the pH is different from the isoelectric point from the protein, the non-covalent interactions will be stronger than when the isoelectric point is achieved (Rodriguez Patino & Pilosof, 2011). The

results are a greater interaction between these molecules with multiple benefits like better emulsification capacity and capsules that are more resistant to environmental conditions, which is desirable for the food industry (Serfert et al., 2013). On the other hand, the tri-dimensional profile of the protein may be modified by the pH, which may reduce it solubility near the isoelectric point, changing their emulsifying properties (Wihodo & Moraru, 2013). This is undesirable for the *spray drying* process since it is required that the wall material must be water soluble (Labuschagne, 2018).

Although proteins are well described as good encapsulating materials, their use in association with other materials is still studied (Dordevic et al., 2014) aiming to understand the impact of its use on the formation of polymers in the microencapsulation of bioactive compounds (Labuschagne, 2018). The use of proteins on encapsulation create an effect of preferential migration of protein to the droplet-air interface and formation of a high-protein-content film on the particle surface (Tontul & Topuz, 2017). Once this film is converted into a glassy skin with high glass transition temperature when subjected to hot air inside the dryer, the interactions of the particles with the chamber of spray-dryer is decreased, resulting in the increase in product yield (Bhusari, Muzaffar, & Kumar, 2014). By this way, the two main proteins used on encapsulation purpose are discussed below.

### *2.3.1 Skim milk powder and whey protein isolate*

Skim milk powder is the most common protein-based wall material used for microencapsulation because of its high efficiency with low cost and easy access (Cano-higuita et al., 2013) and is composed by lactose and proteins (mainly caseins) (Aghbashlo, Mobli, Madadlou, & Rafiee, 2013). The protein portion have an excellent amphiphilic character, offering the optimal characteristics for microencapsulation of bioactive compounds while the carbohydrate portion offer a low glass transition temperature (Jarunglumlert, Nakagawa, & Adachi, 2015). This amphiphilic character may be due their ability to rapidly absorb the hydrophobic compounds forming thick layers with low lateral mobility that could be bounded with other protein complex by cohesive bonds, preventing the coalescence or flocculation of the emulsion by electrostatic and/or steric bonds (Shamaei et al., 2017).

The whey protein isolate (WPI) is a by-product of the cheese industry and does have multiples applications on the food industry (Pinto et al., 2015). It is a mixture of different proteins as β-lactoglobulin, α-lactalbumin and other minor constituents like serum albumin and immunoglobulins (Lam & Nickerson, 2015). The whey protein can be used alone or in combination with other wall materials, and presents an excellent ability of film formation and great retention capability of nutrients in encapsulation process were explored by the literature (Bazaria & Kumar, 2016; Bhusari et al., 2014; Shishir & Chen, 2017).

The interaction of the WPI and the oils can occur by hydrophobic interactions between triacylglycerol and the less polar sites of the proteins (Encina, Vergara, Giménez, Oyarzún-Ampuero, & Robert, 2016). It could react promoting changes on the microenvironment on these proteins, partially changing their tertiary structure and characteristics like increasing the polarity around the tryptophan and tyrosine amino acids, creating less polar zones that are free to interact with low polar molecules (W. Liu et al., 2016).

*2.3.2 Soy protein isolate (SPI)*

Actually, there is a growing trend in the substitution of animal proteins for plant proteins in order to develop less allergenic products, promoting a reduction in health risks due to non-exposure to allergens (Tontul et al., 2016). The soy protein is the main vegetable protein used as wall material, probably due to its commercial availability (F. Liu, Chen, & Tang, 2014). It has good encapsulating properties like film formation and emulsifying abilities, which is desirable for encapsulation of bioactive compounds and other oil or water-soluble compounds (Muzaffar & Kumar, 2015). These properties do reflect on the water activity resistance from the microcapsules and, by consequence, their ability to preserve its polymeric structure and protect the core, even on high water activities (above 0,70) (Rascón et al., 2011). Since proteins are molecules with high biological value, the use of soy proteins microcapsules can improve the nutritional value of food products, especially when it comes to flavors and bioactive compounds encapsulation (F. Liu et al., 2014).

Studies show that SPI can act in different ways according to the core characteristics. With hydrophobic bioactives like essential oils or curcumin, the SPI interact by hydrophobic interactions, promoting a migration effect of these molecules from the polar sites to less polar sites (F. Liu et al., 2014; Tapal & Tiku, 2012). It also can act as a physical barrier, similar to carbohydrates, entrapping the molecules inside their structure (Alla Nesterenko, Alric, Silvestre, & Durrieu, 2012).

When compared to other legume proteins, SPI has superior encapsulating properties with a greater capacity of retention, although they possess similar emulsifying capacities like the formation of polymer and capacity of dissolution in water (F. Liu et al., 2014). However, although SPI is widely studied in the encapsulation of essential oils, further studies are required on its use to encapsulate of bioactive compounds such as polyphenols.

### *2.3.3 Other protein-based wall materials*

Between the proteins that have possible application or were already applied on encapsulation by *spray drying*, various plant proteins can be found. Pea proteins, wheat proteins, rice proteins, oat proteins, sunflower proteins (Alla Nesterenko et al., 2013), kidney, red and mung beans (F. Liu et al., 2014) are some examples of vegetable proteins that were already used on encapsulation by *spray drying*. Other proteins can be extracted from vegetable wastes, including soy wastes (de Figueiredo, Yamashita, Vanzela, Ida, & Kurozawa, 2018), olive kernel (Roselló-soto, Barba, Parniakov, & Galanakis, 2015) and leaves (Tamayo Tenorio, Boom, & van der Goot, 2017), and must be observed for further applications on *spray drying*.

## 2.4 Blends of wall materials

As mentioned before, in view of encapsulating different types of bioactive compounds, some combinations of two or more encapsulating materials are made, in an attempt to eliminate undesirable characteristics or improve the encapsulation performance. Some of the objectives of creating blends are:

(1) To amplify the retention of volatile compounds: A higher retention of volatile content such as flavors and polyphenols can be achieved if the microcapsule wall

consists of a protein associated with a glycolic element (Calvo, Hernández, Lozano, & González-Gómez, 2010; Moser et al., 2017; Sansone et al., 2011; Soottitantawat et al., 2015). This may be achieved because of the reduction of the glass transition temperature that comes along with the addition of proteins, accelerating the formation of the wall around the core material (Encina et al., 2016).

(2) To upgrade the emulsifying properties of the wall material: Blends made of a carbohydrate-based wall material in association with a protein-based wall material show a higher emulsifying ability and can be used in view of create more stable films and to improve the emulsifying properties of the wall material (Labuschagne, 2018; Alla Nesterenko et al., 2013).

(3) To promote greater resistance to environment and upgrade the shelf-life: While the carbohydrate portion of the wall material protects the core against the oxidation, the protein portion maintain the structure of the microcapsule and may stabilize the core by electrostatic interactions (Alla Nesterenko et al., 2013). Both the oxidation resistance and structure maintenance are desirable for a longer shelf-life (Rodea-González et al., 2012).

## 2.5 Conclusion

Through this review, it is possible to understand that the wall material may act in different ways on the encapsulation. The differences between encapsulation with each wall material do not come only from organic matrices, but also from how these molecules interact with the core or other wall materials, if applied. The use of blends seems to be a viable alternative to amplify the efficiency of the process, widening the range of possibilities of use. Finally, we encourage further studies in this field, especially with regard to new biopolymers, blends formulations and physicochemical characterization of these materials, in order to understand the interactions that may occur between the core, the wall material and/or interactions of the wall material and the medium.

## 2.6 Conflicts of interest

The authors declare that there are no conflicts of interest.

# 3 HYDROALCOHOLIC EXTRACTION, ULTRASOUND-ASSISTED EXTRACTION AND ENCAPSULATION OF POLYPHENOLS FROM VEGETABLE RESIDUES

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**- ORIGINAL ARTICLE -**

## Abstract

Bioactive compounds are biomolecules that have potential role on health maintenance and, there is an increasing interest on the extraction of these from vegetable residues. Various solid-liquid extractions are performed to reach these biomolecules, like solvent extractions and ultrasound-assisted extractions. The aim of this study was to evaluate the extraction of polyphenols compounds applying a multi-step hydroalcoholic extraction and ultrasound-assisted extraction on the Fruit and Vegetables Flour (FVF), with and without enzymatic process, followed by an encapsulation by *spray drying*. The total polyphenol content of the extracts were 76.28 ± 2.17 to 92.32 ± 5.79 mg $GAEg^{-1}$ FVF for HAE and 113.02 ± 2.71 to 134.48 ± 1.66 mg $GAEg^{-1}$ FVF. The microstructure of the FVF had significantly changes post treatment on each extraction type. The UAE was the extraction type that most modified the microstructure of the FVF, increasing the release of bound polyphenols. The capsules had TPC varied from 0.54 ± 0.04 to 1.92 ± 0.04 mg $GAEg^{-1}$ of powder, with spherical and tipped morphology. These data

demonstrates that the FVF have higher antioxidant potential than was previously related on the literature.

**Keywords:** solid-liquid extraction, physical extraction, enzymatic extraction, polyphenols, vegetable residues

## 3.1 Introduction

Bioactive compounds are biomolecules that have potential role on treatment and prevention of human diseases (Miller, Feucht, & Schmid, 2019; Pérez-Jiménez, Elena Díaz-Rubio, & Saura-Calixto, 2015). On the last decades, a growing interest on extracting these compounds from vegetable residues has surged since vegetable waste are a global problem that cost billions of dollars per year to manage while they're also a valuable source of bioactive compounds (Corrêa-Filho, Lourenço, Duarte, Moldão-Martins, & Alves, 2019; Plazzotta, Manzocco, & Nicoli, 2017; Sagar, Pareek, Sharma, Yahia, & Lobo, 2018).

Most of bioactive compounds are molecules produced by secondary metabolism of vegetable cells and may be present as extractable polyphenols (or free polyphenols) or non-extractable polyphenols (or bound polyphenols) (S. Liu et al., 2019; Pinaffi et al., 2020), with the second one forming complexes with starch and non-starch polysaccharides like, cellulose, hemicellulose, and pectin (F. Zhu, 2018), and proteins (S. Liu et al., 2019). The polyphenols are bound on the non-starch polysaccharides structure mostly by hydrogen bounds given by their hydroxyl groups (D. Liu et al., 2019) and hydrophobic interactions on the surface of the polymers of the vegetable or in specific sites on the microstructure of these polymers (Jakobek & Matić, 2019; F. Zhu, 2018).

In order to extract of polyphenols from food systems, different solid-liquid extractions can be performed, like the conventional method of solvent extraction (Pinaffi et al., 2020; F. Zhu, 2018) and other novel technologies like ultrasound-assisted extractions (Balasubramaniam et al., 2019; Maran et al., 2017).

Polyphenols creates hydrophobic interactions with the polymeric structure of vegetables, than the use of organic solvents on the extraction aims to reduce the affinity of the polyphenols with the polymeric structure allowing the extraction of these molecules

and, to extract compounds with different polarities (Acosta-Estrada et al., 2014; F. Zhu, 2018). The polymeric composition of the vegetable matrix is a relevant parameter to observe when extracting bound polyphenols, since they interact in different manners (Jakobek & Matić, 2019).

The interaction of the polyphenols and the polymers can be interrupted with modifications on the polymer structure. On this way, enzymatic treatments play a relevant role on the extraction of bound polyphenols when used as pre-treatments on the matrix in order to modify their microstructure and polymeric composition, allowing higher extraction of bound polyphenols is less time (Gligor et al., 2019; Pinaffi et al., 2020; Rajha et al., 2018).

The vegetable cell structures are also affected by ultrasound for solid-liquid extraction of polyphenols (Acosta-Estrada et al., 2014; Tiwari, 2015). The ultrasound-assisted extraction (UAE) promote a formation of bubbles on the medium and these bubbles are responsible for the cavitation effect on the matrix. Bubbles are disrupted when make contact with the vegetable cell structures, creating the mechanical effect that breaks the polymers and create the micro pores, allowing the extraction of the polyphenols (Medina-Torres, Ayora-Talavera, Espinosa-Andrews, Sánchez-Contreras, & Pacheco, 2017; W. Wang et al., 2017). Exposure time, sample:extractor proportion and applied potency (in Watts) can reduce the yield of the polyphenol extraction by degradation of these molecules (Maran et al., 2017).

A lot of the extraction studies present one single extraction of the polyphenols of the matrix which means an waste of bound polyphenols varying from 24% to 85% of total polyphenol content according to the vegetable matrix (Acosta-Estrada et al., 2014; Gligor et al., 2019; Pinaffi et al., 2020; Zhou et al., 2017). Considering that the complexity of vegetable polymers and the type of ligation with polyphenols impact the extraction capacity, it is suggested that multiple extraction cycles can increase the polyphenol extraction. To improve the extraction of free polyphenols from the vegetable matrix, was applied three consecutive cycles of conventional extraction with 80% aqueous acetone (Yang, Dang, & Fan, 2018), also, three cycles of UAE on passion fruit rind (C. G. de Souza, Rodrigues, e Silva, Ribeiro, & de Brito, 2018).

Since the polyphenols are sensible to ambient conditions, an encapsulation process, like *spray drying*, is recommended (Desai, Haware, Basavaraj, & Murthy, 2019; Kiritsakis, Goula, Adamopoulos, & Gerasopoulos, 2018; Rezende, Nogueira, & Narain, 2018; Xue, Su, Meng, & Guo, 2019). In this process, compounds that are sensitive to environmental conditions can be encapsulated with aid of a wall material, giving to the physicochemical properties from the selected wall material a high importance on the process (Mohammed, Tan, Manap, Alhelli, & Hussin, 2017; Negrão-Murakami et al., 2017). For this purpose, maltodextrin is frequently used as wall material (Araujo-Díaz et al., 2017; Shamaei et al., 2017; Tolun, Altintas, & Artik, 2016) due to its resistance to great water activity variations (Araujo-Díaz et al., 2017) and the ability to prevent oxidation by reducing the oxygen permeability (Labuschagne, 2018; Shishir, Xie, Sun, Zheng, & Chen, 2018).

Following the tendency of waste valorization, Ferreira *et al* (2013) characterized a Fruit and Vegetables Flour (FVF) made of the solid waste obtained in the production of an isotonic drink (Ferreira et al., 2013; Martins, Chiapetta, Paula, & Gonçalves, 2011). This drink uses selecta orange (*Citrus sinensis*), passion fruit (*Passiflora edulis*), watermelon (*Citrullus lanatus*), lettuce (*Lactuca sativa*), courgette (*Cucúrbita pepo*), carrot (*Daucus carota*), spinach (*Spinacea oleracea*), mint (*Mentha s.p.*), taro (*Colocasia esculenta*), cucumber (*Cucumis sativus*) and rocket (*Eruca sativa*) as ingredients. The FVF final composition have 26% of carbohydrates, 9.5% of proteins, 5% of lipids, 11,1% of moisture and ash (Ferreira et al., 2013). Also, 48.4% of the biopolymers present on the FVF are dietary fiber as cellulose, hemicellulose, soluble lignin, insoluble lignin and resistant starch, where 80% are insoluble fiber (Brito, Carrajola, Gonçalves, Martelli-Tosi, & Ferreira, 2019). The FVF presented 22.49 ± 1.59 mg $GAEg^{-1}$ of total polyphenol content (TPC), and the hydroalcoholic extraction with 75% ethanol solution was the best extraction type in terms of total polyphenol content for a single extraction of polyphenols on this matrix (Santos & Gonçalves, 2016). In addition, 88 compounds were also identified (28 were phenolic acids, 32 flavonoids and 28 other polyphenols), with the hesperidin as the most abundant compound (Gonçalves et al., 2018).

The aim of this study was to evaluate the extraction of polyphenols compounds applying a multi-step hydroalcoholic extraction and ultrasound-assisted extraction on the FVF, with and without enzymatic process, followed by an encapsulation by *spray drying*.

## 3.2 Methodology

The Fruit and Vegetables Flour (FVF) was used as matrix on this study.

### *3.2.1 Extraction process:*

#### *3.2.1.1 Hydroalcoholic extraction (HAE)*

This extraction was performed with ethanol 75% as extractor at 40°C for 24 hours. The supernant was recovered and the residue was submitted to a new extraction with equal volume of extractor. The process was considered finished when the total phenolic compounds of the extract did not presented statistical difference ($P < 0.05$) on three consecutive extractions (Figure 1).

Three different conditions were applied as follows, with solid:liquid ratio on 1:15 (m:v). The Viscozyme® multi enzymatic complex was acquired from Sigma Aldrich and have FBGU/G ≥100 Betaglucanase units of enzymatic activity with 1.2gcm$^3$ of density.

I. FVF : ETOH (1g:15mL);
II. FVF : Viscozyme® : ETOH [(1g): 25µL*;15mL)]
III. FVF : PC**: Viscozyme® : ETOH [(1g:1g) 25µL*;30mL)]

* - Added on each cycle. ** - *In natura* pineapple crown (PC) was cut in pieces with 0.5cm$^2$. Also, a blank was made with the pineapple crown treated as the condition II for the polyphenol extraction. Each condition was made on triplicate.

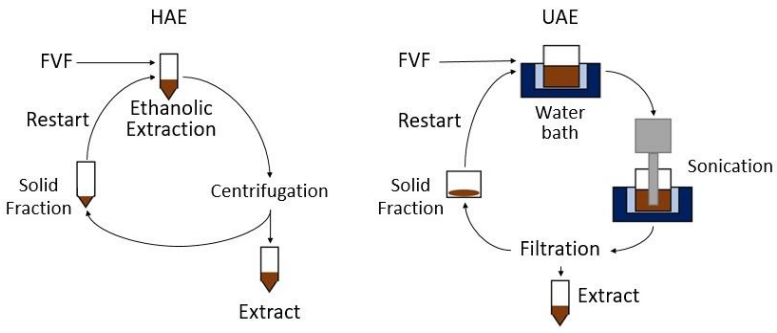


**Figure 1:** Scheme of each extraction cycle.

*3.2.1.2 Ultrasound-assisted extraction (UAE)*

Sample was put in a NT232 Dubnoff water bath (Novatécnica, Brazil) at 30°C for 30 minutes. After that, the solution was exposed continuously to ultrasonic waves (Maran et al., 2017) on UIP1000hdT Ultrasound (1000W/20kHz, Hielscher, Germany), adjusted for 500w and 100% amplitude (Widyasanti, Halimah, & Rohdiana, 2018). The process occurred for 8 minutes and limiting temperature of 60°C (Moorthy et al., 2017), using an ice bath. After filtered the supernatant was recovered. On the sequence, the residue was submitted to a new extraction with equal volume of extractor. The process was considered finished when the total phenolic compounds of the extract did not presented statistical difference ($P < 0.05$) on three consecutive extractions (Figure 1). Three different conditions were applied as mentioned above.

*3.2.1.3 Total polyphenol content (TPC)*

The total polyphenol content was made by Folin-Ciocalteu technique (Singleton, Orthofer, & Raventós-Lamuela, 1999) on each supernatant recovered from the cycles of each extraction process. The polyphenol determination was made on Victor Nivo Microplate Reader (Perkin Elmer, German). The results expressed as mg of Gallic Acid Equivalent per gram of sample (mg $GAEg^{-1}$) and was made in five replicates.

*3.2.1.4 Scanning Electron Microscopy (SEM) of the residues*

The FVF residues of each cycle and each condition were oven dried (60ºC) and observed on a Tabletop Scanning Electron Microscope TM3000 (Hitachi, Japan). They were not coated with gold and were observed on six random points on 15kV (Bernardo, Ascheri, Chávez, & Carvalho, 2018).

*3.2.2 Encapsulation of FVF extracts by spray-dried*

*3.2.2.1 Feed solution preparation (FD)*

The solution to be encapsulated (SC) was obtained from mix of all volume of each extraction cycle. The final cycle was considered the first of three cycles did not presented statistical difference ($P < 0.05$) TPC amount (Table 1). For the encapsulation, 80mL of

SC and 160mL of distilled water where homogenized and maltodextrin 10DE (Corn Products, Brazil) was added until the solution reaches 28°Brix (FD).

**Table 1:** Number of cycles and correspondent total volume of extract.

| Samples | Cycles used | SC (mL) |
|---|---|---|
| HAE.I | 6 | 270 |
| HAE.II | 7 | 315 |
| HAE.III | 6 | 540 |
| UAE.I | 6 | 900 |
| UAE.II | 5 | 750 |
| UAE.III | 5 | 1500 |

*3.2.2.2 Encapsulation process*

The solutions were dried on a mini spray Dryer Büchi B-190 (Büchi, Switzerland) in following conditions: 60mBar vacuum, 75lb/pol$^2$ of pressure, 170°C of inlet temperature and 90°C of outlet temperature (V. B. De Souza, Thomazini, Balieiro, & Fávaro-Trindade, 2015). The powders were stored on laminated bags at ambient conditions. The powder yield was achieved as described by De Sá Mendes *et al* (2019) (de Sá Mendes et al., 2019). Physical properties (moisture, microstructure and density) were determined in triplicate, while the total polyphenol content was in quintuplicate.

*3.2.2.2.1 Moisture*

The moisture was determined on an Infrared Moisture Analyzer IV2000 (Gehaka, Brazil) (Bernal, Ramos, & Baena, 2019).

3.2.2.2.2 Density determination

The bulk density (Bd) and the tapped density (Td) were determined according to De Sá Mendes *et al* (2019) (de Sá Mendes et al., 2019) technique. Also, the flowability [Carr index (CI)] and cohesiveness [Hausner Correlation (HC)] were achieved as described by Jedlinska & Samborska (2018) (Jedlińska et al., 2018). The powders flowability was considered as follows: CI < 15%: great flowability; CI between 15% and 20%: good flowability; CI between 20% and 35%: intermediary flowability; CI between 35% and 45%: low flowability; CI > 45%: very low flowability. On the other hand, their

cohesiveness was considered as follows: HC < 1.2: low cohesiveness; HC between 1.2 and 1.4: intermediary cohesiveness; HC > 1.5: high cohesiveness.

3.2.2.2.3 Powders polyphenol content (PPC)

A 10% solution (m/v) of each powder obtained was made and the total polyphenol content was achieved by Folin-Ciocalteu technique (Singleton et al., 1999). The polyphenol determination was made on Victor Nivo Microplate Reader (Perkin Elmer, German). The results expressed as mg of Gallic Acid Equivalent per gram of sample (mg $GAEg^{-1}$) and was made in five replicates.

3.2.2.2.4 Scanning Electron Microscopy (SEM) of powders

The powders obtained from each condition were observed on a Tabletop Scanning Electron Microscope TM3000 (Hitachi, Japan). They were not coated with gold and were observed on six random points on 15kV (Bernardo et al., 2018).

### *3.2.3 Statistical analysis*

The results were expressed by mean ± standard deviation and were submitted to ANOVA and Tukey test (Microsoft Excel Statistics). Results were considered significant with a 95% confidence level ($P < 0.05$).

## 3.3 Results

### *3.3.1 Extraction process*

As stablished, the extraction cycles were continued until the accumulated total polyphenol content did not present statistical difference ($P < 0.05$) on three consecutive cycles, then a total number of eight cycles were necessary for conditions HAE.I and HAE.III and nine cycles for HAE.II (Figure 2.A).

The HAE conditions had overall a similar behavior on the increase of the polyphenols on the extract until the $5^{th}$ cycle. After that, the Viscozyme® effect on the polymeric structure can be seen on HAE.II, creating significant difference from conditions I and III that remained until the last cycle.

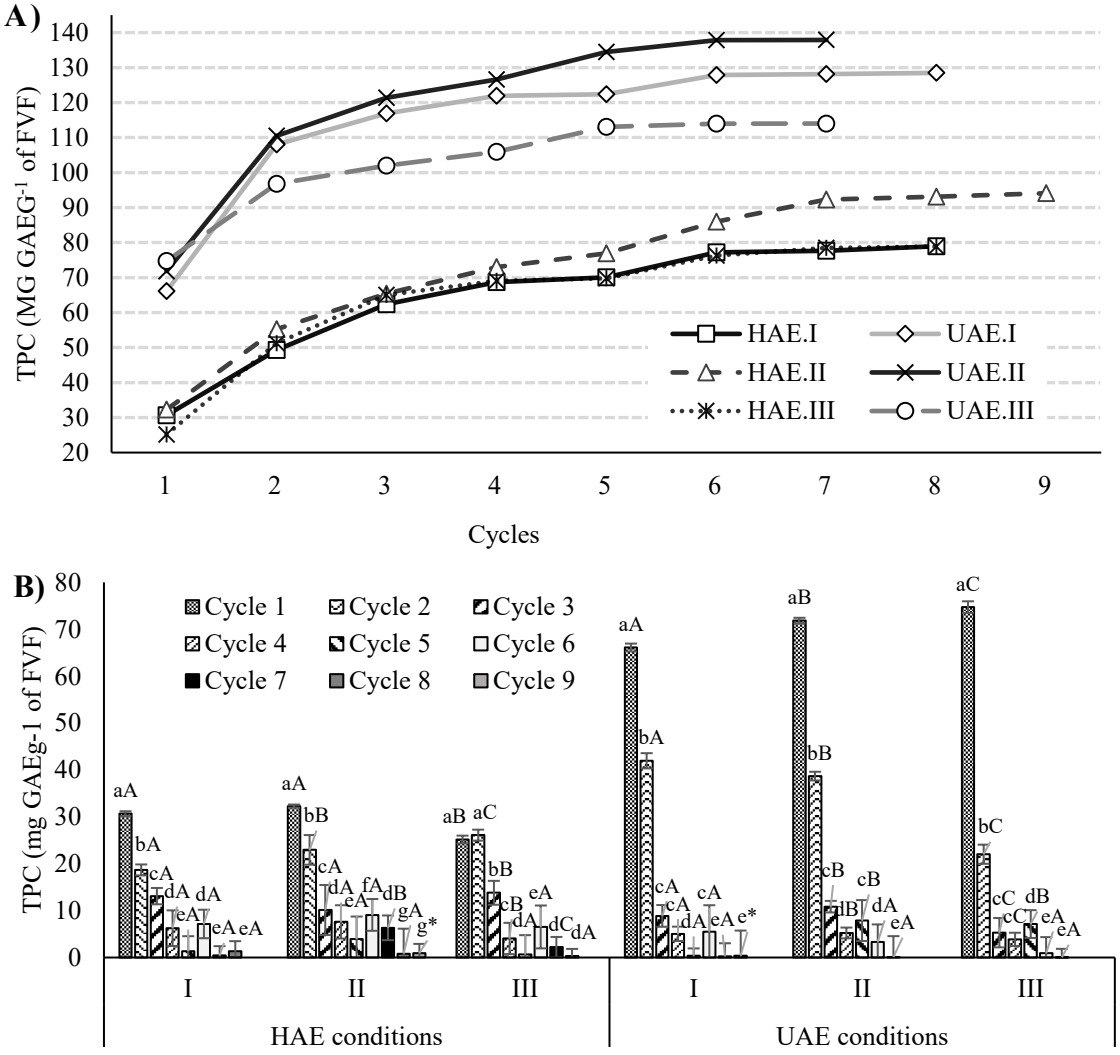


**Figure 2:** Total polyphenol content (mg GAEg$^{-1}$ FVF) on different extraction cycles of FVF by hydroalcoholic extraction [HAE - 75% (v/v)] and ultrasound-assisted extraction [UAE], on three different conditions (I, II, III) with solid:liquid ratio on 1:15 (m:v): A) Behavior on the extractions cycles (n = 3). B) TPC from each cycle and condition (n = 3). Lowercase letters means significant difference between polyphenol content on each cycle on the same condition and extraction type ($P < 0.05$). Capital letters means significant difference between the same cycles between the conditions of the same extraction type ($P < 0.05$). *Signalized cycles were executed only on the respective condition. I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

The increase of the extraction capacity of the condition II can be explained by the compatibility of Viscozyme® (enzyme complex that includes cellulases, hemicellulases, pectinases) (de Figueiredo et al., 2018) and the FVF matrix (48.4% of the biopolymers are dietary fiber as cellulose, hemicellulose, soluble lignin, insoluble lignin and resistant starch) (Brito et al., 2019), demonstrating that this enzyme complex can be used to improve extraction of polyphenols (Rajha et al., 2018; Zhou et al., 2017). These results

are in accordance with the present literature that reports increases on the extraction capacity of polyphenols with enzymatic treatments (Rajha et al., 2018; Waterhouse, Sun-Waterhouse, Su, Zhao, & Zhao, 2017b).

Comparing the condition I with the condition II (Figure 2.A), the enzymatic treatment on HAE was much more significant than on UAE in terms of increase of extraction capacity. However, when condition I is compared with condition III, the condition III had lower effect on TPC (HAE.III). It may be related with the enzyme concentration on this condition, since the enzyme:substrate ratio is a relevant parameter to observe on enzymatic treatments (Heemann, Heemann, Kalegari, Spier, & Santin, 2019). The pineapple crown also have relevant proportions of cellulose, hemicellulose and lignin (de C.M. Miranda et al., 2019) and could act as a competitive substrate for Viscozyme®. Viscozyme® was kept as $25\mu L.g^{-1}$ of FVF on the condition II, but on the condition III the proportion of enzyme to total solids goes to half. This effect can reduce the potential of extraction (Gligor et al., 2019; Swer, Mukhim, Bashir, & Chauhan, 2018). Also, other studies with Viscozyme® suggest the use of higher concentration of enzyme:substrate in a range of 6% to 7.5% (v/w) (Antunes-Ricardo et al., 2018; Mackėla, Andriekus, & Venskutonis, 2017).

The HAE profile of extraction (Figure 2.B) demonstrates a gradual decrease of the extraction capacity as the number of cycles increase and was similar in all conditions. It may be related with the reduction of free polyphenols on FVF composition. However, on the $6^{th}$ cycle the extraction had a significant increase in all conditions, compared with the $5^{th}$ cycle. It could be related with the long exposure to controlled temperature, since heating may weaken the hydrogen bonds between polyphenols and the polymeric structures of vegetables, promoting the release of these molecules (Mokrani & Madani, 2016; Ozdal, Capanoglu, & Altay, 2013; Pinaffi et al., 2020).

On UAE, eight cycles were necessary on condition I, and seven for conditions II and III (Figure 2.A). All conditions had similar behavior on the extraction. However, the total phenolic content of the UAE.III was significantly lower than the others. It may be related with the presence of pineapple crown structural polymers. The pineapple crown cellulose, hemicellulose and lignin probably acted as a physical barrier for the bubbles

created on the medium by the ultrasound (W. Wang et al., 2017), reducing the efficiency of the process.

The UAE conditions had similar profile of extraction (Figure 2.B). After the 2nd cycle, a significant reduction on the extraction capacity of each cycle was observed, followed by a gradual decrease on the extraction capacity. This behavior could be related with the high degree of cell disruption on the FVF promoted by the acoustic cavitation process on the 1st and 2nd cycles already, allowing the higher concentration of polyphenol to be extracted on these cycles (Maran et al., 2017; Medina-Torres et al., 2017). The extraction profile of condition III had different behavior. UAE.III presents the 1st cycle extracting more efficiently than the UAE.I and UAE.II 1st cycles, but with a significant reduce of the capacity of the 2nd cycle. It may be related with the particle size of the pineapple crown applied on the FVF. The higher particle size reduces the surface area exposed to cavitation, decreasing its effect (Azmir et al., 2013; Medina-Torres et al., 2017).

It is possible to observe that the cycles 1 to 3 are the most relevant cycles, extracting between 80% (HAE) to 90% (UAE) (Figure 2.A), while the cycles 4 to 6 had gradual decreases on the extraction capacity (Figure 2.B).

In a previous study, the FVF presented total polyphenol content of 22.49 ± 1.59 mg $GAEg^{-1}$ of flour (Santos & Gonçalves, 2016), these data demonstrates that on HAE most of the antioxidant potential of the matrix is not explored with one single extraction (cycle 1), with approximately 60-70% of the total polyphenol content remaining on the polymeric matrix of FVF. On UAE, after one single extraction (cycle 1), almost 50% of the total polyphenol content remains bounded on the FVF matrix. However, this is a relevant side finding since it demonstrates that the FVF have a highly content of

antioxidant dietary fibers on the matrix, which represents approximately 48% of total mass of FVF (Brito et al., 2019).

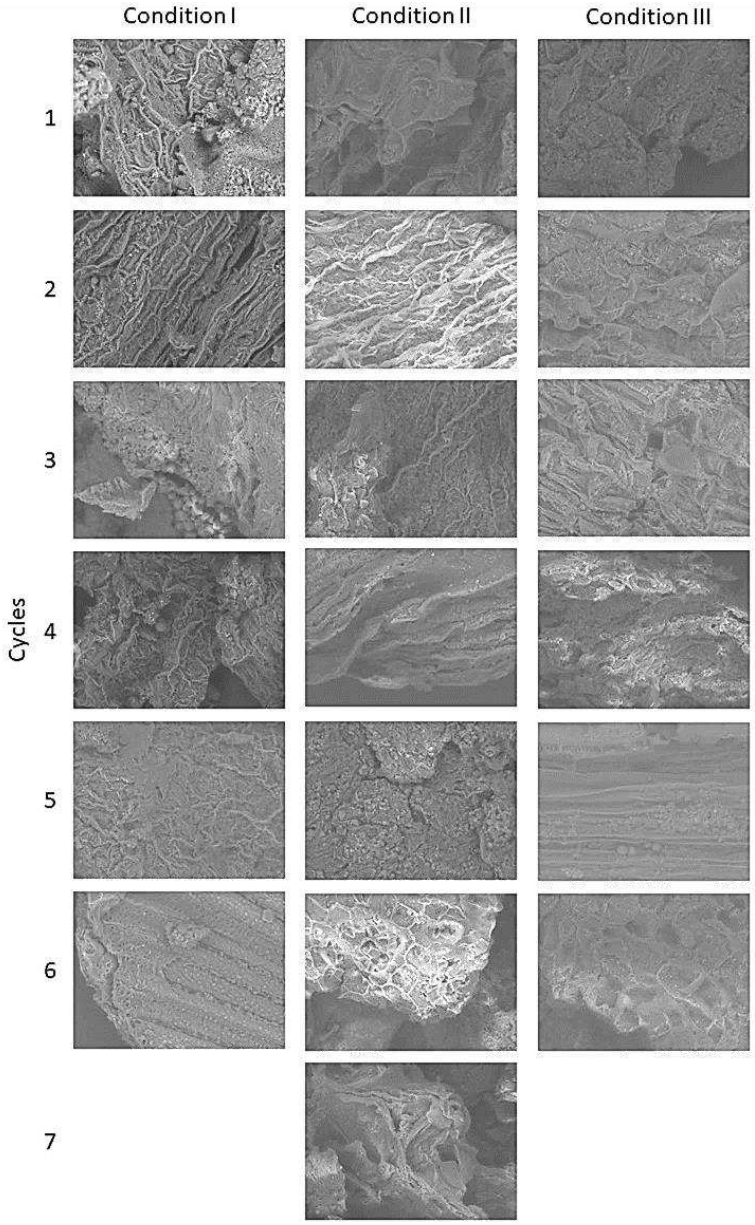


**Figure 3:** SEM images from hydroalcoholic extraction [HAE -75% (v/v)] residues on three different conditions (I, II, III) considering as the final cycle the first of three cycles that did not presented statistical difference ($P < 0.05$) on TPC amount. Magnifications at 1800x. I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

When comparing the HAE SEM images (Figure 3) with the native microstructure SEM images of FVF (Figure 4), it is possible to indicate that even the ethanolic medium at extraction temperature could actively modify the cell wall structures of FVF. The medium and temperature gradually modifies the FVF microstructure, increasing the bioactives extraction (F. Zhu, 2018). Even if the FVF has a relevant protein content (Ferreira et al., 2013), the presence of natural bromelain did not promoted a significantly change on the microstructure of the FVF (Figure 3, Condition III) as all conditions presented similar SEM images. This is an expected result since cellulose, hemicellulose and lignin are the main structural polymers of FVF (Brito et al., 2019).

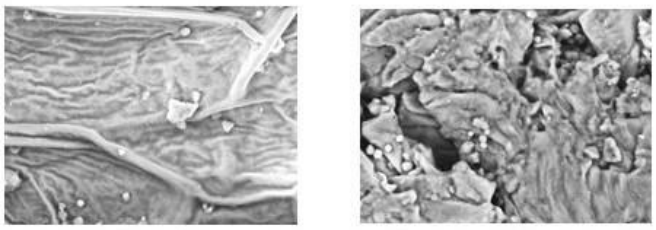

**Figure 4:** SEM images of FVF native microstructure. Magnifications at 1000x.

The UAE SEM images (Figure 5) demonstrates great difference from the FVF native microstructure (Figure 4) due the acoustic cavitation process, creating the pores on the FVF (Maran et al., 2017; Medina-Torres et al., 2017). The pores are formed on the beginning of the extraction process (Medina-Torres et al., 2017) and remained on the samples until the final cycles. However, the condition III seems to be less affected by the others, which reinforces the theory of the protective effect of the pineapple crown against the cavitation effect.

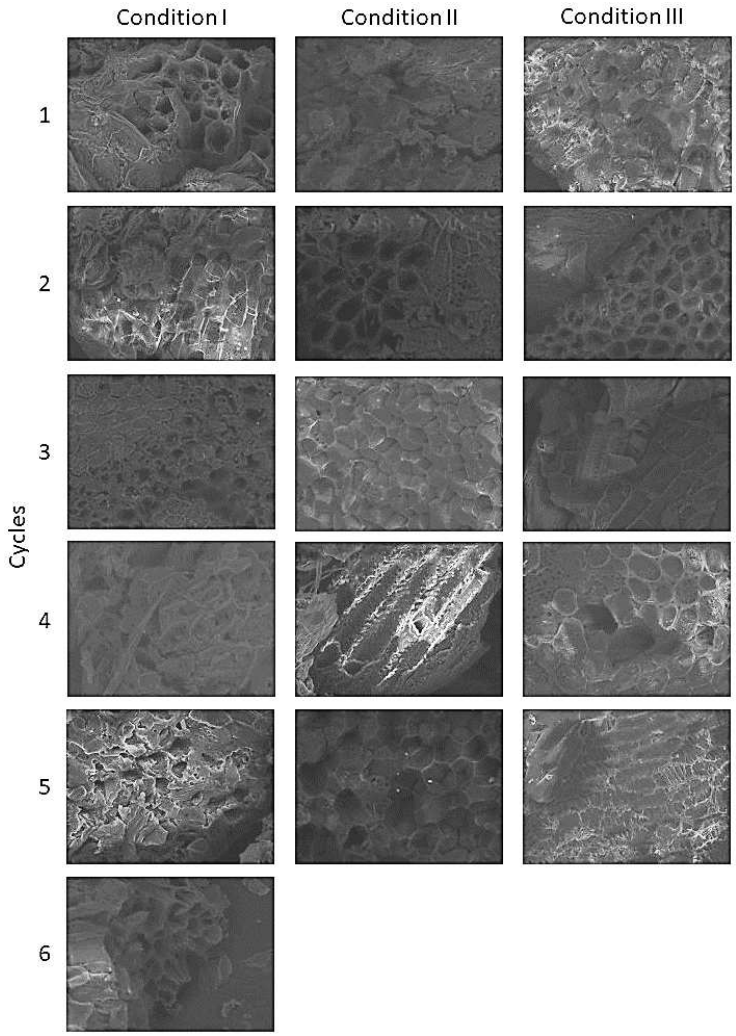


**Figure 5:** SEM images from ultrasound-assisted extraction [UAE] residues on three different conditions (I, II, III) considering as the final cycle the first of three cycles that did not presented statistical difference ($P < 0.05$) on TPC amount. Magnifications at 1800x. I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

Considering as the final cycle the first of three cycles that did not presented statistical difference ($P < 0.05$) on TPC amount, it was possible to observe that the enzymatic treatment with Viscozyme® was efficient on HAE and irrelevant for UAE, when compared with the condition I (Table 2).

**Table 2:** Total polyphenol content on FVF extracts from hydroalcoholic extraction [HAE - 75% (v/v)] and ultrasound-assisted extraction (UAE) on three different conditions (I, II, III) considering as the final cycle the first of three cycles that did not presented statistical difference ($P < 0.05$) on TPC amount (n = 3).

| Sample Conditions | HAE | | UAE | |
|---|---|---|---|---|
| | TPC (mg GAEg$^{-1}$ FVF) | Cycles | TPC (mg GAEg$^{-1}$ FVF) | Cycles |
| I | $77.19 \pm 0.93^{aA}$ | 6 | $127.86 \pm 4.09^{aB}$ | 6 |
| II | $92.32 \pm 5.79^{bA}$ | 7 | $134.48 \pm 1.66^{bB}$ | 5 |
| III | $76.28 \pm 2.17^{aA}$ | 6 | $113.02 \pm 2.71^{cB}$ | 5 |

Lowercase letters on each column means difference between the conditions on the same extraction type ($P < 0.05$). Capital letters means difference between the extraction types on the same condition ($P < 0.05$). I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

Overall, the UAE was more efficient than the HAE on the extraction of the polyphenols from FVF considering that 3 cycles on UAE can extract higher content than 9 cycles of HAE, respecting the conditions. Also, the best extraction condition was the UAE.I since it demands no enzymatic treatment and still had higher total polyphenol content than any HAE condition. However, HAE is simpler and cheaper, since only the extractor solution and a controlled heating equipment are required to perform it, while UAE requires specific equipment with sample cooling adjustment to perform it properly.

Although HAE is a simpler method of extraction, the FVF extracts had a higher concentration of polyphenols than tomato peel extracts with 38.78 ± 0.05 mg GAEg$^{-1}$ (Grassino et al., 2019) and wine shoot wastes with 32.1 ± 0.9 mg GAEg$^{-1}$ (Moreira et al., 2018) or even chokeberry polyphenols with 27.8 mg GAEg$^{-1}$ (Ćujić et al., 2016), peach with 36.3 mg GAEg$^{-1}$ (Mokrani & Madani, 2016), and Calville White Winter apple cultivar with 36.07 ± 0.19 mg GAEg$^{-1}$ (Morresi et al., 2018). The UAE extracts also had higher concentration of polyphenols than other ultrasound-extracts like: olive kernel with 60.75 ± 0.40 mg GAEg$^{-1}$ (Roselló-Soto et al., 2015), *Nephelium lappaceum* L. fruit peel extracts with 55.26 ± 0.16 mg GAEg$^{-1}$ (Maran et al., 2017) and Persian lime (*Citrus latifolia*) wastes with 58.13 ± 0.4 mg GAEg$^{-1}$ (Medina-Torres et al., 2019). This demonstrates FVF as a polyphenol source since they could be easily extracted by simple and cheaper methods.

*3.3.2 Encapsulation process*

The powder yield is an important factor to observe on the *spray drying* process (Hoskin, Xiong, & Lila, 2019). On this study, it varied from 63.56% to 79.36% (Table 3) and was in accordance with other powders made with vegetable waste extracts as core and maltodextrin as wall material as the literature reports values as 72.81% to 76.49% for olive leaves extracts (Kiritsakis et al., 2018), 20.14% to 48.20% for grape pomace extracts (Tsali & Goula, 2018). The difference on the yield values could be related with the extraction type and condition applied, since the drying parameters were maintained constant (de Sá Mendes et al., 2019; Tontul & Topuz, 2017).

The density is an important factor to observe for storage. All conditions and treatments had intermediary to high cohesiveness as well as low to intermediary flowability (Table 3). The differences between the bulk density and the tapped density reveals that capsules had low rate between the core material and the wall material (Stranzinger et al., 2019).

The encapsulation process was proportional to the extraction efficiency from each extraction process and condition. Considering all steps included on each condition and extraction type, the best powder obtained was UAE.I, since it has similar polyphenol content as the UAE.II with no need of enzymatic treatment and was obtained in much less time than the HAE. However, HAE capsules still had satisfactory polyphenol content, since they are similar to the literature. The powder polyphenol content of the capsules obtained is in accordance with other powders obtained from vegetable wastes that were considered with high antioxidant activity, as a powder of citrus by-products conventional extracts that had $1.66 \pm 0.02$ mg $GAEg^{-1}$ powder (Papoutsis et al., 2018) and $1.69 \pm 0.01$ mg $GAEg^{-1}$ powder for red pepper wastes conventional extract powder (Vulić et al., 2019).

**Table 3:** Powder yield, polyphenol content and physical properties of spray dried powders obtained from hydroalcoholic extraction [HAE - 75% (v/v)] and ultrasound-assisted extraction (UAE) on three different conditions (I, II, III) (n = 3).

| Parameters | HAE | | | UAE | | |
|---|---|---|---|---|---|---|
| | I | II | III | I | II | III |
| Powder Yield (%) | 79.36 | 68.89 | 78.00 | 78.91 | 63.56 | 72.63 |
| Moisture (%) | $5.57 \pm 0.35^{aA}$ | $6.5 \pm 0.17^{bA}$ | $5.67 \pm 0.58^{aA}$ | $5.20 \pm 0.44^{aA}$ | $4.47 \pm 0.12^{bB}$ | $4.63 \pm 0.32^{bB}$ |
| Bulk density ($g/cm^3$) | $0.257 \pm 0.013^{aA}$ | $0.304 \pm 0.026^{bA}$ | $0.300 \pm 0.017^{bA}$ | $0.401 \pm 0.037^{aB}$ | $0.381 \pm 0.009^{aB}$ | $0.367 \pm 0.016^{bB}$ |
| Tapped Density ($g/cm^3$) | $0.441 \pm 0.003^{aA}$ | $0.471 \pm 0.005^{bA}$ | $0.396 \pm 0.004^{cA}$ | $0.802 \pm 0.073^{aB}$ | $0.584 \pm 0.025^{bB}$ | $0.647 \pm 0.046^{cB}$ |
| CI % | $41.62 \pm 2.60^{aA}$ | $35.32 \pm 5.97^{bA}$ | $24.22 \pm 4.73^{cA}$ | $47.16 \pm 2.62^{aA}$ | $34.81 \pm 1.28^{bA}$ | $43.21 \pm 2.14^{aB}$ |
| CI classification | Low flowabillity | Low flowabillity | Intermediary flowabillity | Low flowabillity | Intermediary flowabillity | Low flowabillity |
| HR | $1.72 \pm 0.08^{aA}$ | $1.55 \pm 0.14^{bA}$ | $1.32 \pm 0.09^{bA}$ | $1.90 \pm 0.10^{aA}$ | $1.53 \pm 0.03^{bA}$ | $1.76 \pm 0.06^{aA}$ |
| HR classification | Highly cohesive | Highly cohesive | Intermediary cohesiveness | Highly cohesive | Highly cohesive | Highly cohesive |
| Polyphenol content (mg $GAEg^{-1}$ powder) | $1.00 \pm 0.04^{aA}$ | $1.23 \pm 0.03^{bA}$ | $0.54 \pm 0.04^{cA}$ | $1.50 \pm 0.16^{aB}$ | $1.92 \pm 0.04^{bB}$ | $0.62 \pm 0.04^{cB}$ |

Lowercase letters means difference between the conditions on the same extraction type ($P < 0.05$). Capital letters means difference between the extraction types on the same condition ($P < 0.05$). I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

All conditions produced capsules with spherical shape, tipped morphology different sizes (Figure 6) and similar moisture contents, as expected from spray dried powders (Chávez Montes et al., 2019; Corrêa-Filho et al., 2019; Laokuldilok & Kanha, 2015; Oliveira et al., 2018; Papoutsis et al., 2018). In addition, all capsules obtained showed no cracks, indicating a good encapsulation process since it can reduces the air permeability, preserving the core during the storage (Castel et al., 2018; Desai et al., 2019; Shamaei et al., 2017).

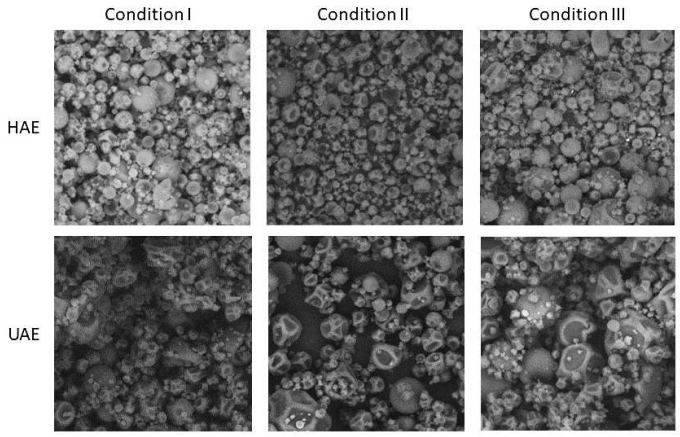


**Figure 6:** SEM images from spray dried powders obtained from hydroalcoholic extraction [HAE -75% (v/v)] and ultrasound-assisted extraction (UAE) on three different conditions (I, II, III). Magnifications at 1800x. I – FVF:ETOH (1g:15mL); II – FVF:Viscozyme®:ETOH (1g:25µL:15mL); III – FVF:PC:Viscozyme®:ETOH [(1g:1g):25µL:30mL].

### 3.4 Final considerations

FVF is the best choice as supplement, compared powder of extracts. It not only have high antioxidant potential as demonstrated on this study as well as its macronutrients compositions favors health, especially by the antioxidant dietary fiber content (Andrade, Ferreira, & Gonçalves, 2014; Brito et al., 2019; Ferreira et al., 2013; Jakobek & Matić, 2019; S. Liu et al., 2019; Santos & Gonçalves, 2016; F. Zhu, 2018).

The powders obtained by the *spray drying* process can be used as food products ingredient since they are easier to be storage and added on food products formulation (de Sá Mendes et al., 2019; Tolve et al., 2016). Additionally, as the maltodextrin was used as wall material, their benefits to food industry includes high water solubility, low viscosity and bland flavor, which are desirable for food processing (Saavedra-Leos et al., 2015).

Also, it is related that antioxidant powders are desirable to enrich food products (Desai et al., 2019; Sagar et al., 2018; Wilkowska, Ambroziak, Adamiec, & Czyżowska, 2016)

### 3.5 Conclusion

The enzymatic treatment applied with Viscozyme® was only relevant on HAE, allowing an increase of almost 15% on total polyphenol extraction, but it was not on UAE. The use of the pineapple crown was not favorable for the extraction of polyphenols from FVF. FVF and encapsulation of the extracted FVF polyphenols indicated that both samples has potential as functional ingredient.

### 3.6 Acknowledgments

The authors thanks the Embrapa for the collaboration with this study.

### 3.7 Conflicts of interest

The authors declare that there are no conflicts of interest.

## 4 FINANCIAMENTO

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