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System dynamics and product quality during fluidized bed agglomeration of phytochemical compositions

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ABSTRACT

Pressure fluctuation signals linked to growth kinetics and fluiddynamic data were used to characterize system dynamics during fluidized bed encapsulation/agglomeration of phytochemical compositions using microcrystalline cellulose of two different sizes as seed particles. Two distinct operating modes were investigated: intermittent (interrupting the atomization of the feed composition for a specific period of time in order to reduce the system humidity) and continuous (without interruption the atomization of the feed). The increase in the agglomeration percentage to a certain value during the processes caused an increase in the standard deviation of the amplitude of the pressure fluctuation signals while the system remained stable, indicative of slight changes on solids circulation patterns. However, the standard deviation of the amplitude of the pressure fluctuation signals tended to decrease when agglomeration percentage reached a value able to affect significantly the system stability, in which agglomerates growth tended to cause the system collapse. This behavior was most evident for small size seed particles and continuous operating mode. The changes in the standard deviation of the amplitude of pressure fluctuation signals show strong evidence that this method would be able to detect alterations in system dynamics and can be a useful tool for process control and system monitoring.

1. Introduction

Encapsulation is an efficient process to stabilize, avoid degradation and control the release rate of herbal bioactive compounds, being of great importance on the development of nutraceuticals and functional food products.

Fluidized bed encapsulation/agglomeration is a promising alternative technology for encapsulation of herbal extract based compositions, generating granules with improved physicochemical properties adequate for post processing operations in pharmaceutical, food and agricultural industries [1].

The process consists of spraying a composition of the bioactive herbal compounds entrapped in an encapsulating matrix over a fluidizing bed of seed particles. The composition impinges and spread on the particles surface and the fluidization air evaporates the solvent resulting in coated/agglomerated granules [1,2]. Granules growth can occur by coalescence of two or more particles, termed agglomeration, or by surface layering or coating of single particles [3]. It is a complex process where drying, coating and agglomeration of the encapsulating matrix containing bioactive compounds occur at the same time in one single step suffering the action of many processing variables. The predominance of particle growth by agglomeration or coating depends on the operating conditions used and also on the physicochemical properties of the seed particles and feed compositions.

However, fluidized bed process can be impaired due to the formation of large agglomerates due to high humidity in the system and particle stickiness, leading to alterations in solid circulation and gas flow patterns causing the system defluidization or bed collapse. Therefore, the fast detection of emergence of system instabilities, through transitions on particle flow regimes based on in-line process measurements, would be a useful tool to guarantee safe operational conditions in order to obtain a product with the desired properties.

Pressure fluctuations signals have been reported as a significant indicator of fluidization dynamics [4]. They are commonly measured with the use of sensitive high frequency pressure transducers connected to a pressure probe either attached to the inner wall of the column or inserted into the fluidized bed [4]. The simplicity of measuring pressure fluctuations as compared to other parameters, leads to its widespread application in fluidization research [5].

Time-series signals of pressure fluctuations in a gas–solid fluidized bed can be analyzed in different ways like, statistical, frequency and chaos or state space analysis [5]. Statistical analysis is a common method in time domain, where the amplitude of signals of pressure, expressed as standard deviation or variance is studied to identify the regime changes from bubbling to turbulent regime [6]. The analysis in the frequency domain is typically carried out using Fast Fourier Transform (FFT) to obtain the power spectral density (PSD) that is an appropriate

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qualitative analysis to physically determine the fluidization behavior [5]. A good review of the use of time-series analyses of pressure fluctuation in gas–solid fluidized bed is presented by van Ommen et al. [7].

In this study, the feasibility of the statistical analysis of fluidized bed pressure fluctuation signals linked to growth kinetics data to characterize changes in system dynamics during the fluidized bed encapsulation/agglomeration of phytochemical compositions was investigated. Microcrystalline cellulose (Celphere®) of two different sizes was used as seed particles, and two distinct operating modes were evaluated: intermittent (interrupting the feed composition atomization for a specific period of time in order to reduce the system humidity) and continuous (without interruption of feed atomization).

Pressure fluctuation signals were measured in the system wall at a frequency of 200 Hz for a period of 2 min. Growth kinetics was evaluated by the determination of granule sizes from samples of particle withdraw during the agglomeration process. Fluidized bed performance was evaluated based on coating efficiency and percentage of agglomeration.

Rosmarinus officinalis L. (Labiatae) was used as model plant species for preparation of feed composition. This plant species, commonly known as rosemary, is an aromatic herb native to Mediterranean countries, and cultivated worldwide. This plant is used as a flavor, beverage, herbal medicine and in cosmetic products. It possesses confirmed antimicrobial, antifungal, anti-ulcerogenic, anti-inflammatory, and antioxidant activities and is a potential food preservative [8,9].

This work aims to correlate fluidized bed pressure fluctuation analysis with the events that occur during the encapsulation/agglomeration process, from the atomization of the feed composition, the adhesion in the seed particles and the consequent granule growth, and product properties.

2. Materials and method

2.1. Materials

Dried leaves of *R. officinalis* L. (Labiatae) were purchased from Santosflora (Mairiporã, SP, Brazil). The leaves were comminuted in a knife mill (model MA-680, Marconi, Piracicaba, SP, Brazil) until they passed through an 800 μm sieve (mean diameter of 300 μm).

The encapsulating compositions were based on Poloxamer® 407 (Plucare F127, Via Farma, São Paulo, Brazil), stearic acid (Via Farma, São Paulo, Brazil), maltodextrin (MOR-REX 1910 — Corn Products of Brazil, Mogi Guaçu, SP, Brazil) and Acacia gum (Fibregum B®, NEXIRA, São Paulo, Brazil).

Pellets of microcrystalline cellulose Celphere® (Asahi Kasei Chemicals, Japan), namely CP 507 (size ranging from 500 to 710 μm) and CP 305 (size ranging from 300 to 500 μm), were used as seed particles in the fluidization experiments. The seed particles were classified as type B, according to Geldard's classification [10].

2.2. Experimental procedure

2.2.1. Extraction of bioactive compounds

Dried and powdered leaves of *R. officinalis* L. (Labiatae) were placed in contact with ethanol 70% (*v*/*v*) in a proportion of 0.10 kg plant/L of solvent under dynamic maceration. The extraction was performed in a jacketed stirred vessel under mechanical stirring for 60 min at a controlled temperature of 50 °C [11]. The resulting extract was filtered in a vacuum filtration system and concentrated in a rotary evaporator (vacuum pressure of 500 mm Hg at 50 °C) until a solid content of 10.1 ± 0.22% *w*/*w*. The concentrated extract presented density of 0.94 g/cm^3, measured with a pycnometer.

2.2.2. Characterization of the seed particles

The seed particles were characterized by determining the mean particle diameter, particle size distribution, SPAN, shape factor and flow properties.

The mean particle diameters and particle size distributions were determined via image analysis. Scaled images of the particles were acquired using a Canon Power Shot SX50 HS digital camera (12.1 megapixels) and analyzed using an Image Analysis System (Image-Pro Plus® 7.0).

The SPAN of size distribution was determined according to Eq. (1) [12].

$$\text{SPAN} = \frac{d_{90} - d_{10}}{d_{50}} \tag{1}$$

where, d_{10}, d_{50} and d_{90} are the particle diameters for which, respectively, 10, 50 and 90% of the particles are smaller.

The particles shape factor (Φ) was determined by measuring three characteristic dimensions of a certain number of particles with the aid of the Image Analysis System (Image-Pro Plus® 7.0). The shape factor was defined as the ratio between the smallest divided by the largest particle diameters:

$$\varphi = \frac{\text{minor diameter}}{\text{largest diameter}} \tag{2}$$

The Hausner ratio (FH) and Carr index (IC) were the flow properties and compressibility characteristics determined for the seeds and agglomerated product. FH and IC were estimated from particle density measurements by Eqs. (3) and (4), respectively [13].

$$\text{FH} = \frac{\rho T,1250}{\rho b} \tag{3}$$

$$\text{IC} = \frac{\rho T,1250 - \rho b}{\rho T,1250} x100 \tag{4}$$

where, ρb is the bulk particle density (ρb = m/v); and ρT,1250 is the tapped bulk particle density, determined by dividing the mass by the volume occupied by the particles after tapping the probe 1250 times from a distance of 14 mm using a Caleva® Tapped Density Tester TDT.

The fluid dynamic characterization of the seed particles (dry bed) was also performed in order to determine the experimental values of the minimum fluidization velocity (U_{mf}). Fluid dynamic tests were performed for the fluid bed system loaded with 300 and 600 g of each seed particle at room temperature (25 °C). Data of bed pressure drop as a function of the gas velocity fed to the bed were obtained. The velocity determined by the intersections of the lines connecting the fixed bed pressure drop and the ones at fluidized bed conditions were considered the minimum fluidization velocity, U_{mf} [14].

2.2.3. Preparation of the encapsulating compositions

Encapsulating composition containing the concentrated rosemary extractive solution added with the encapsulating carriers was prepared at a solid content of 25% (*w*/*w*). The concentrated herbal extract/encapsulating materials ratio was 1:5 (*w*/*w* — dry base) and the encapsulating blend was: Poloxamer 407 (20%), stearic acid (10%), maltodextrin DE10 (30%) and Arabic gum (40%). Poloxamer 407 and stearic acid were mixed and melted at 70 °C. The concentrated herbal extract was blended with the mixture of Acacia gum and maltodextrin DE10 heated at 70 °C, previously dissolved in purified water and hydrated for 24 h. This aqueous phase was then dispersed in the lipid phase and vigorously stirred at 18,000 rpm for 5 min using an ultra Turrax (T18, IKA-USA). Although, the system can be classified as O/W dispersion, its structure is highly heterogeneous when observed by optical microscopy, consequence of the complex mixture composition. Density of the composition was determined in a pycnometer and viscosity value was obtained using a Brookfield rheometer LV-DVIII (coaxial-cylinder geometry) with a SC4-25 spindle at a temperature of 30 ± 2 °C, at a specific shear rate in rotational speed of 140 rpm.

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