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PROCESSING OF *Rosmarinus officinalis* LINNE EXTRACT ON SPRAY AND SPOUTED BED DRYERS

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Abstract - This article presents an investigation of the potential of spray and spouted bed technology for the production of dried extracts of *Rosmarinus officinalis* Linné, popularly known as *rosemary*. The extractive solution was characterized by loss on drying, extractable matter and total phenolic and flavonoid compounds (chemical markers). The product was characterized by determination of loss on drying, size distribution, morphology, flow properties and thermal degradation and thermal behavior. The spray and spouted bed dryer performance were assessed through estimation of thermal efficiency, product accumulation and product recovery. The parameters studied were the inlet temperature of the spouting gas (80 and 150°C) and the feed mass flow rate of concentrated extract relative to the evaporation capacity of the dryer, W_s/W_{max} (15 to 75%). The atomizing air flow rate was maintained at 20 l/min with a pressure of 196.1 kPa. The spouting gas flow rate used in the drying runs was 40% higher than the gas flow under the condition of minimum spouting. The spray drying gas flow rate was fixed at 0.0118 kg/s. Under the conditions studied, performance in the spray and spouted bed drying of *rosemary* extract was poor, causing high degradation of the marker compounds (mainly the phenolic compounds). Thus, process improvements are required before use on an industrial scale.

Keywords: Drying; Spouted bed; Spray drying; Dried extract; Medicinal plants; Rosemary.

INTRODUCTION

Recently, there has been a tremendous surge in the world demand for herbal or medicinal plant products. According to a World Health Organization survey, about 70-80% of the world populations rely on nonconventional medicine in their primary healthcare. This strategy is mostly based on the use of medicinal plant products know as botanicals, herbal medicines or phytomedicines (Akerele, 1993; Calixto, 2000; Chan, 2003).

Basically, the standardized herbal preparations are marketed in the form of liquid, viscous preparations and also as powders resulting from the drying and comminuting of plants or from the drying of an extract. The advantages of the dried extract over conventional liquid forms are lower storage costs and higher concentration and stability of active substances.

In essence, herbal dried extracts are produced through the drying of a concentrated extractive solution from herbal materials (leaves, roots, seeds, whole plant, inflorescence, etc.), resulting in a dried powder. Several drying techniques can be utilized including freeze-drying, spray drying and spouted bed drying (Teixeira, 1996; Souza, 1997; Senna et al., 1997; Cordeiro, 2000; Runha, 2000; Runha et al., 2001; Bott, 2001; Souza, 2003; Souza and Oliveira, 2005; Cordeiro and Oliveira, 2005), with spray dryers being the most commonly used in the herbal

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*To whom correspondence should be addressed processing industries. Nevertheless, the operating conditions used in the drying and thermal processing of bioproducts like dried extracts and herbal materials might have a considerable impact on the properties and cost of the product, generating different degrees of loss of active compounds (Souza and Oliveira, 2005; Raghavan and Orsat, 1998). Therefore, the development of studies to elucidate the effects of the drying processes as well as operating conditions on system performance and product properties during the manufacture of standardized dried extracts is fully justified.

In this work, an evaluation of dryer performance and the physical and chemical product properties during manufacture of spray and spouted bed dried extracts of *Rosmarinus officinalis* Linné, popularly known as *rosemary*, is presented.

Rosemary (*Rosmarinus officinalis* Linné) is a common household plant, grown in many parts of the world. It is used for flavoring food and as a beverage, as well as in cosmetics. In folk medicine it is used as an antispasmodic in renal colic and dysmenorrheal, in relieving respiratory disorders and in stimulating the growth of hair. Rosemary extract relaxes the smooth muscles of the trachea and intestine and has choleretic, hepatoprotective and antitumerogenic activity (Al Sereiti et al., 1999; Newall et al., 2002; Albu et al., 2004). These attributes can be related to its high content of phenolic compounds like caffeic acid derivatives such as rosmarinic acid. Phenolic compounds are secondary plant metabolites that have long been associated with flavor and color characteristics of fruits and vegetables. These compounds attract great interest due to their postulated health-protecting properties, foremost of their antioxidative effect, manifested by the ability to scavenge free radicals or to prevent oxidation of low-density lipoprotein (Newall et al., 2002; Miliauskas et al., 2004; Albu et al., 2004). However, adequate intakes and rates of absorption of phenolic compounds would be necessary to obtain these beneficial effects (Andlauer et al., 2003).

MATERIALS AND METHODS

Material

Dried and powdered *Rosmarinus officinalis* leaves with a mean diameter of 0.3 mm were used in the preparation of the extractive solution. This material was purchased from Oficina de Ervas Pharmacy, located in Ribeirão Preto, State of São Paulo, Brazil, and characterized by determination of loss on drying (measurement of the moisture plus volatile content), extractable matter and concentration of the marker compounds (total flavonoid and phenolic compounds) used in the optimization of the extraction conditions and in degradation studies during drying.

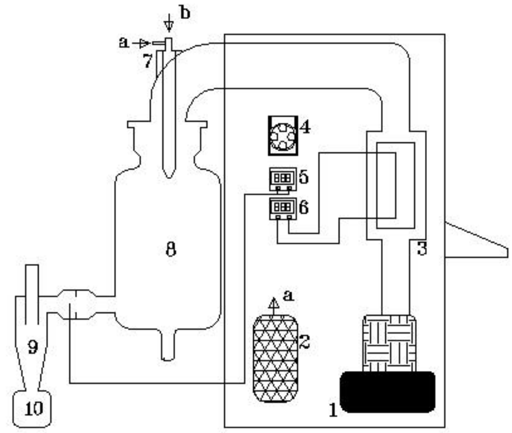
Ethyl acetate, methanol, acetone, aluminum chloride, ethanol and chloridric acid and o-phosphoric acid were acquired from LabSynth. Hexametilenetetramine, sodium tungstate, and phosphomolibidic acid (Vetec); dehydrated quercetin and gallic acid (Sigma-Aldrich); colloidal silicon dioxide (Tixosil® 333, Rhodia) and maltodextrin DE 14 (Mor Rex® 1914, Corn Products Brazil) were used as reagents and standard materials.

Spray and Spouted Bed Dryers

A bench-top spray dryer, model SD-05 (Lab-Plant, U.K.) with a concurrent flow regime was used in this work. The drying chamber had a diameter of 215 mm and a height of 500 mm. The main components of the system were the extract feed system, composed of a peristaltic pump, a two-fluid atomizer (inlet orifice diameter of 0.5 mm) and an air compressor; a feed system for the drying gas, composed of a blower and an air filter; and a temperature control system for the drying gas. The dried product was collected in a Lapple cyclone with a diameter of 0.085 m (cut diameter of 3.9 μm). Figure 1 contains a schematic diagram of the spray dryer used.

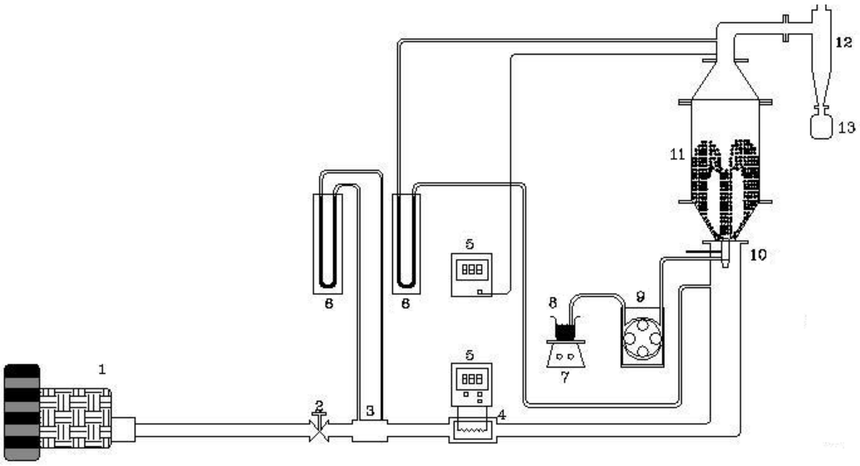
The spouted bed dryer consisted of a conical base with an internal angle of 40° and an inlet orifice diameter of 33 mm. A cylindrical column with a diameter of 150 mm and a height of 400 mm was connected to the conical base of the dryer. The upper part of the equipment was formed of another cone and by a cyclone. All parts were made of stainless steel. Figure 2 shows a schematic diagram of the experimental rig. Teflon® beads of concave-cylindrical shape with a mean diameter of 5.45 mm, a shape factor of 0.96, a specific surface of 5.27 cm^2/g and a density of 2.16 g/cm^3 were used as inert material. Teflon was selected due to its inert nature, high thermal stability, low friction coefficient, insolubility and lack of toxicological effects. The main components of the system were a 7.5 HP blower, a flow meter, an electric heater (power of 5000 W) and a temperature controller. The extract feed system consisted of a double fluid atomizer with internal mixing (0.8 mm hole), a peristaltic pump and an air compressor. The dried product was collected in a stainless steel Lapple-type cyclone with a diameter of 0.095 m and a cut diameter of 4.1 μm (for the experimental conditions used). Thermocouples, pressure transducers and a thermohygrometer were employed in the equipment instrumentation.

The other devices used were an analytical balance (Mettler Tolledo AG204, Switzerland), a vacuum pump, a drying and sterilization oven (Fanem Model 315 SE – Brazil), a vacuum filtration system, a rotary evaporator, mechanical and magnetic stirrers (Fisatom model 713 and 753 – Brazil), a Traceable® thermohygrometer, a digital thermometer (Minipa APPA MT-520), a centrifuge (Fanem Excelsa II, Model 206 BL – Brazil), a HP 8453 UV-VIS spectrophotometer running the HP Chem-Station® software, a Zeiss DSM 960 scanning electron microscope, and an Olympus® BX60MIV optical microscope connected to an Image Pro-plus® 4.5 image analysis system.



1. Blower + air filter
2. Air compressor
3. Heater
4. Peristaltic pump
5. Temperature controller
6. Inlet thermocouple
7. Atomizer: (a) compressed air; (b) feed suspension
8. Drying chamber
9. Cyclone
10. Dry product collector

Figure 1: Schematic diagram of the spray dryer used.



1. Blower
2. Valve
3. Orifice plate
4. Air heater
5. PID control
6. Manometer
7. Stirrer
8. Concentrated extract
9. Peristaltic pump
10. Atomizer
11. Spouted bed
12. Cyclone
13. Dried extract

Figure 2: Schematic diagram of the experimental rig.

Experimental Methods

a) Evaporation Capacity of the Dryers, W_{max}

The evaporation capacity of the spray and spouted bed dryers, W_{max}, as a function of feed flow rate and of inlet temperature of the drying air was determined. These results are important for selection of the ranges of feed flow rate of the extract during the spray drying runs. The methodology and results on the evaporation capacity of this spouted bed and spray drying were presented elsewhere (Souza and Oliveira, 2002, 2004).

b) Development of the Extraction Procedure

A 2^3 factorial design was used to evaluate the effect of the extraction conditions on extraction efficiency (Montgomery, 1978; Box et al., 1978), aiming at selecting the condition providing the extract with maximum solids concentration and total phenol and flavonoid contents. The parameters studied were extraction temperature (30 and 50°C); plant-to-solvent mass ratio, m_{pl}/m_{sol}, (0.1 and 0.2) and extraction time (1 and 2 h). Ethanol:water (70:30 in weight) was the extraction solvent. Water and ethanol were used as solvents to facilitate extraction of the polar and nonpolar substances.

The extractive solution was prepared in an extraction system composed of a jacketed stirred vessel connected to a heating circulating batch with temperature control (Brookfield TC-500). After extraction, the crude extract was filtered in a vacuum system using filter paper (grade 80G) and concentrated three times in a rotary evaporator at a temperature of 50ºC and a vacuum pressure of 650 mm Hg, and its density, solids concentration and total flavonoid and phenol contents were determined. An analysis of variance (ANOVA) was carried out on the experimental results aiming at selecting the conditions for extract preparation. ANOVA results show that extraction time did not have a significant effect on the extractive process. The variable m_{pl}/m_{sol} had a significant effect on total flavonoid and on solids content at a significance level of 0.05. The effect of the variable m_{pl}/m_{sol} on total phenol content was significant at a α level of 0.10. Temperature was only statistically significant for total phenol content ($\alpha \leq 0.05$). Thus, the following conditions were selected for preparation of the extractive solution: an extraction time of 1 hour; an extraction temperature of 50°C and a plant-to-solvent mass ratio of 0.2.

c) Manufacture of the Dried Extracts

The concentrated extractive solution prepared under optimized conditions was added with the drying carriers before drying. A mixture of colloidal silicon dioxide (Tixosil 333®) and maltodextrin DE 14 at a proportion of 40:20 in relation to the total solids content was selected as drying carrier. This selection was based on several preliminary studies.

The drying operation started with injection of the drying air into the spouted bed (SB) previously loaded with Teflon® beads and into the SD-05 spray dryer (SD). The air was heated to the desired temperature and then the concentrated extracts were fed in at a preset flow rate together with the atomizing air. The parameters studied were inlet gas temperature, T_{gi} (80 and 150°C), and the ratio of the mass feed flow rate of the concentrated extract to the evaporation capacity of the dryer, W_s/W_{max} (15 to 75%). The mass flow rate of the drying gas was maintained at 0.0340 kg/s, corresponding to a Q/Q_{ms} ratio of 1.4 for the spouted bed and 0.0118 kg/s for the spray drying. The atomizing air flow rate was maintained at 20 l/min with a pressure of 196.1 kPa. Table 1 shows the variables and operating conditions used in the drying runs.

Measurements of the outlet gas temperature, T_{go}, were taken at regular intervals in order to detect the moment when the dryers attained the steady state (± 15 minutes). Once the steady state was attained, samples of the dried extract were removed (± 35 minutes) and stored in amber bottles for the subsequent analysis. The samples were used for physical and chemical characterization of the dried product. To evaluate systems performance, the inlet and outlet spouting gas temperature, the mass of product accumulated on the inert material and in the drying chamber and the product collected by the cyclone were measured during the operation.

Table 1: Variables and operating conditions used in the drying runs.

Dryers	Processing parameters T_{gi} (°C)	W_s/W_{max} (%)	W_s (g/min)
Spray dryer	80	15	1.8
	80	45	2.8
	80	75	5.8
	150	15	3.3
	150	45	9.9
	150	75	19.0
Spouted bed	80	15	6.0
	80	45	18.0
	80	75	30.0
	150	15	10.0
	150	45	33.0
	150	75	49.0

T_{gi}= inlet gas temperature (°C)
W_s= mass feed flow rate of the concentrated extract (g/min)

d) Physicochemical Properties of the Herbal Material, Extractive Solution and Dried Extracts

The experimental methodologies used for physical and chemical characterization of the herbal raw material, the extractive solutions and the dried product are presented in Table 2.

The total flavonoid content was evaluated by spectrophotometry (UV-vis), using predefined sample mass. The procedure includes hydrolysis of the glycosides, followed by flavonoid extraction with ethyl acetate and color development with a solution of aluminum chloride (Kulevanova et al., 2000). Absorbance was measured at a wavelength of 420 nm after 30 minutes. The percentage of total flavonoid (T_F) is expressed as quercetin (average of three measurements) with a correlation coefficient of 0.9993.

Quantification of the total phenol content was based on the Folin-Denis method. This procedure is based on reduction of the phosphomolybdic-phosphotungstic acid by the phenolic compounds in a basic medium, producing a dark blue color measured at a wavelength of 760 nm. To correlate the measured absorbance with total phenol concentration, a calibration curve was constructed using gallic acid as standard substance with a correlation coefficient of 0.997.

e) Dryer Performance

Spray and spouted bed dryer performances were evaluated through determination of the product recovery ratio, R, and the accumulation rate in the spouted bed, Ac, estimated by mass balance in the dryer, and through evaluation of the energetic efficiency of the system following the methodology presented by Souza (2003). Table 3 contains the equations used in these determinations.

Table 2: Physical and chemical properties of the herbal raw material, extractive solution and dried product.

Properties	Method
Loss on drying , U_P (% g/g)	Gravimetric method, 102 ±1°C (WHO 1998)
Extractable matter, T_E (% g/g)	$T_E = \frac{m\ 500}{p}$ (Teixeira, 1996)
Total flavonoid content, T_F (% g/g)	$T_F = \frac{A\ Fd}{71.716\ C_s}$ (Schiavetto, 2004)
Total phenol content, T_P (% g/g)	$T_P = \frac{A\ Fd}{157.961\ C_s}$ (Schiavetto, 2004)
Flavonoid degradation ratio, D_F (%)	$D_F = \frac{T_{F,ext} - T_{F,d}}{T_{F,ext}}\ 100$
Phenol degradation ratio, D_P (%)	$D_P = \frac{T_{F,ext} - T_{F,d}}{T_{F,ext}}\ 100$
Powder morphology, size distribution and flow properties	Optical microscope with image analysis system (50 times); morphology by S.E.M.; flow properties (loosely packed and tapped bulk densities, Hausner ratio and compressibility index) in accordance with Prista et al. (1995)

Table 3: Equations used in the determination of dryer performance.

Parameter	Equation	Unit
Product recovery ratio, R	$R(\%) = \frac{Mc\,(1\text{-}U_P)}{W_s\ C_s\ \theta}\ 100$	(% kg/kg)
Accumulation ratio, Ac	$Ac(\%) = \frac{(M_f\text{-}M_i)\,(1\text{-}U_p)}{W_s\ C_s\ \theta}\ 100$	(% kg/kg)
Energetic efficiency, η	$\eta = \frac{W_s\ (1 - C_s - C_s\ U_p)\ \lambda}{W_g\ C_{pg}\ (T_{gi} - T_{go})}$	(-)

RESULTS AND DISCUSSION

The herbal raw material had a moisture content (loss on drying) of 10.4 ± 0.2 (% kg/kg), extractable matter of 22.2 ± 0.01 (% kg/kg) and a total flavonoid content of 0.332 ± 0.02 (% kg/kg). The concentrated extract had a density of 0.98 ± 0.01 (g/cm^3), a solids content of 7.32 ± 0.06 (% kg/kg), a total flavonoid content of 11.87 ± 0.02 (mg/g, dry basis) and a total phenol concentration of 146.07 ± 0.23 (mg/g, dry basis). A low value of alcohol content, measured with an alcoholmeter, was obtained (≤ 5%). The concentrated extracts were standardized to a solids content of 11.3% with the addition of the drying carriers before the spray and spouted bed drying runs. During the experiments, samples of the powdered product were taken and used for the physical and chemical characterization of the dried extract. In this characterization loss on drying of the dried extract (≈ residual moisture content), the degradation ratio of the total flavonoid and phenolic compounds, the product size distribution, the particle morphology and the flow properties (loosely packed bulk density, ρ_b; tapped bulk density, ρ_{bt}; real density, ρ_r; compressibility index, IC; and Hausner ratio, HR) were quantified.

The experimental results on the total flavonoid and phenol degradation ratios as a function of the process parameters for spray and spouted bed drying are presented in Table 4. An analysis of variance for the experimental results presented in Table 2 shows that the total flavonoid degradation ratio tends to increase with W_s/W_{max} and T_{gi} for both spray and spouted bed drying. In general, the increase in drying gas temperature resulted in a reduction in total phenol concentration (higher degradation ratio), whereas the parameter W_s/W_{max} had a insignificant effect. However, high degradation ratios were observed for both total flavonoid (13.1 to 49.9%) and total phenol contents (45 to 53%), independent of the drying process used. The phenolic compounds (including the flavonoids) are comprised of a large number of organic molecules with heterogeneous structure. Phenolic substances have an important role as antioxidants, being highly reactive with O_2. Thus, the degradation of phenolic substances, including flavonoids, may be associated with the occurrence of oxidative condensation or decomposition of thermolabile compounds induced by heat and also with the possibility of losses of essential oil and other volatile substances during processing (Blanco et al., 2002).

Table 4: Flavonoid and phenol degradation ratio as a function of processing conditions.

	T_{gi} (°C)	W_s/W_{max} (%)	D_F (%)	D_P (%)
Spray dryer	80	15	22.8	50.7
	80	45	38.3	48.5
	80	75	31.9	45.8
	150	15	26.0	48.3
	150	45	29.6	49.3
	150	75	39.1	48.8
Spouted bed	80	15	13.1	48.2
	80	45	25.0	45.7
	80	75	36.6	45.9
	150	15	32.4	50.2
	150	45	48.6	48.7
	150	75	49.9	53.3

D_F: total flavonoid degradation ratio.
D_P: total phenol degradation ratio.

Loss on drying is an important property of dried extracts, thus being an indicator of drying efficiency. The term "loss on drying" is similar in meaning to moisture content. Nevertheless, since herbal products are a complex mixture of chemical substances, many other volatile substances besides water may be present in the dried product. Thus, loss on drying includes losses of water and other volatile compounds in the samples. This parameter has noteworthy importance in the development of pharmaceutical dosage forms. It has a considerable effect on the chemical and microbiological stability as well as the physical properties of the product, such as size distribution and flow properties.

Figure 3 contains the experimental results of the loss on drying for the spray and spouted bed dried extracts. The dashed line in Figure 3 shows the maximum loss on drying value recommended by the Brazilian Pharmacopoeia (1988) for dried extracts of medicinal plants (≤ 4%). It can be observed that only the spouted bed dried extract obtained at $T_{gi} = 150°C$ and $W_s/W_{max} = 15\%$ falls with in the recommended range.

The size distribution of the powdered extracts was measured by optical microscopy with a 50x magnification and analyzed with version 4.5 of the software Image Pro-plus®. Table 5 contains the experimental results on the mean size diameter obtained for the spray and spouted bed powdered extracts as a function of processing parameters. Both dryers generated polydisperse powder particles within a limited range, with diameters varying from 7 to 22 μm. It can be observed in Table 5 that the spray-dried extracts had mean diameters slightly lower than the spouted bed ones. This behavior may be associated with the physical and chemical properties of the extractive solution fed to the dryers, the atomizing conditions and also fine losses by the cyclone.

The particle morphology of the spray and spouted bed dried extracts was obtained through scanning electronic microscopy (S.E.M.) with a 1000x magnification. Figure 4 shows S.E.M. photomicrographs of the SD and SB dried extracts obtained at $T_{gi} = 150°C$ and $W_s/W_{max} = 15\%$. In Figure 4 a high proportion of irregular and multisized agglomerated particles can be observed for the spouted bed dried extract.

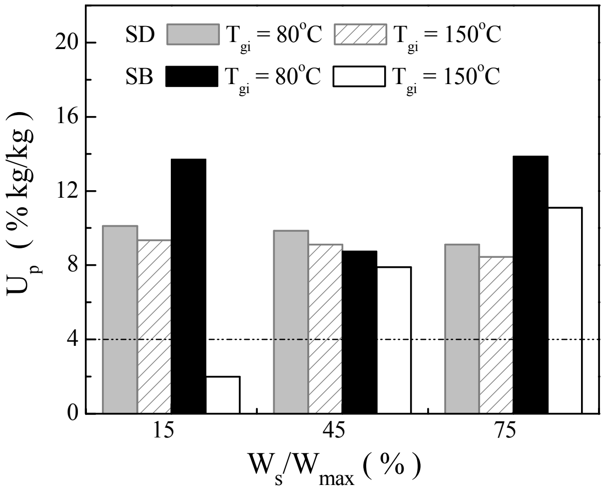
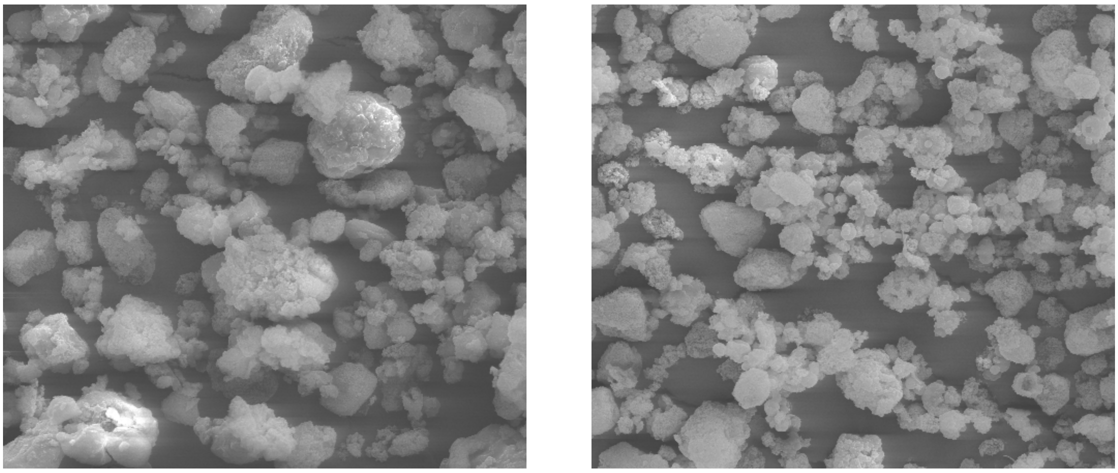


Figure 3: Loss on drying of the spray and spouted bed dried rosemary extracts.

Table 5: Mean size diameter obtained for the spray and spouted bed powdered extracts as a function of processing parameters.

		Equipment	
Variables		**Spray dryer**	**Spouted bed**
T_{gi} (°C)	W_s/W_{max} (%)	$\bar{d}_p$ (μm)	$\bar{d}_p$ (μm)
80	15	9.1	14.6
80	45	7.4	13.5
80	75	10.3	22.3
150	15	9.1	13.0
150	45	12.7	14.8
150	75	13.2	12.4



(a) Spray drying (b) Spouted bed

Figure 4: Typical photomicrographs obtained by S.E.M. for the spray and spouted bed dried rosemary extracts.

The behavior of the powdered product during storage, manipulation and subsequent processing was directly related to its flow properties (loosely packed bulk density, $\rho_{b;}$ tapped bulk density, $\rho_{bt;}$ true density, $\rho_{r;}$ compressibility index, IC; and the Hausner ratio, HR). In general the drying process and operating conditions had a significant effect on these properties. The results reveal that the spray and spouted bed dried extracts obtained in this study did not have good flow properties, with Hausner ratios of 1.98 and 1.82 and compressibility indices of 49 and 44%, respectively. According to Prista et al. (1995) and Jong et al. (1999), Hausner ratios higher than 1.25 are indicative of a cohesive or fairly free flow that is not compressible. On the other hand, to be easily compressed, the IC values should be equal to or lower than 15%. The unfavorable values of HR and IC obtained in this study are associated with the reduced particle size diameter and with the drying carriers used (colloidal silicon dioxide and maltodextrin DE 14).

The spray and spouted bed dried extracts had respectively mean loosely packed bulk densities of 320 ± 10 and 430 ± 10 kg/m^3 and tapped bulk densities of 620 ± 30 and of 780 ± 20, with a real density, measured with a helium picnometer, of 1493.7 ± 2.9 kg/m^3.

Dryer Performance

Spray and spouted bed dryer performances were evaluated through determination of the product recovery ratio, R and the accumulation rate in the spouted bed, Ac, estimated by mass balance in the dryer, and the thermal efficiency of the system. Figure 5 contains the experimental results on the recovery ratio as a function of drying system and operating parameters. Spray drying had higher recovery ratios than spouted bed drying, independent of the operating conditions used. For the spouted bed, the recovery ratio decreased with the increase in W_s/W_{max} ratio, probably due to product accumulation in the bed. These results emphasize the importance of the physical and chemical properties of the composition fed on the overall performance of the drying processes since different results were reported elsewhere (Oliveira et al., 2006).

In order to evaluate product accumulation in the spouted bed, the dryer and the inert material were weighed at the beginning and at the end of the drying. After weighing, pictures of samples of the inert particles were taken in order to record the phenomenon observed (data not shown). At both drying temperatures the spouted bed dryer had a large quantity of product accumulated on the walls, inert materials and cyclone. Figure 6 contains the experimental results on mass accumulated in the spouted bed as a function of operating parameters. The product mass accumulated in the spouted bed increased with the W_s/W_{max} ratio. The accumulation tended to be larger at a temperature of 150ºC, except for W_s/W_{max} = 75%.

Energetic efficiency is a factor commonly used in optimization of the drying processes. In this work this parameter was estimated by energy balance of the drying gas under inlet and outlet conditions. Figure 7 contains the results obtained. As expected, higher values of energetic efficiency were obtained at higher values of the W_s/W_{max} ratio. For a W_s/W_{max} ratio of 15%, higher values of energetic efficiency were obtained at an inlet gas temperature of 80°C. The values of energetic efficiency attained by spouted bed drying were higher than those attained by the spray dryer, showing a better performance with this criterion.

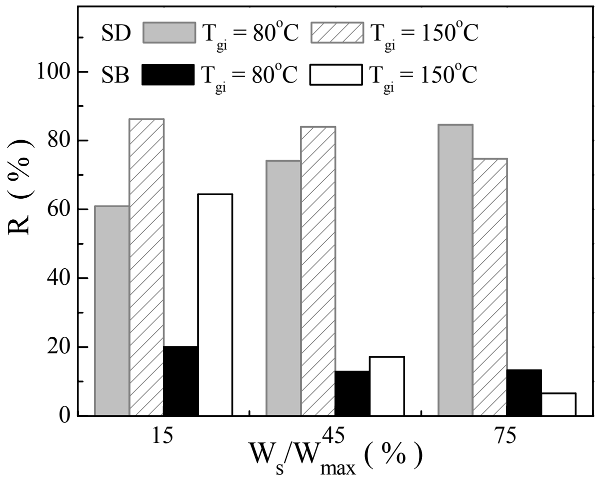


Figure 5: Recovery ratio as a function of drying system and operating parameters.

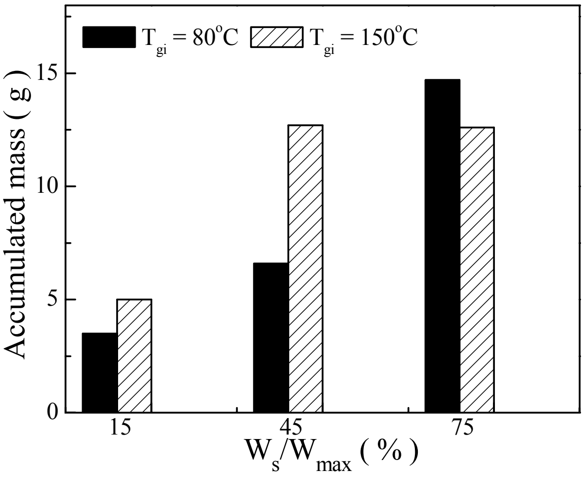


Figure 6: Mass accumulated in the spouted bed as a function of operating parameters.

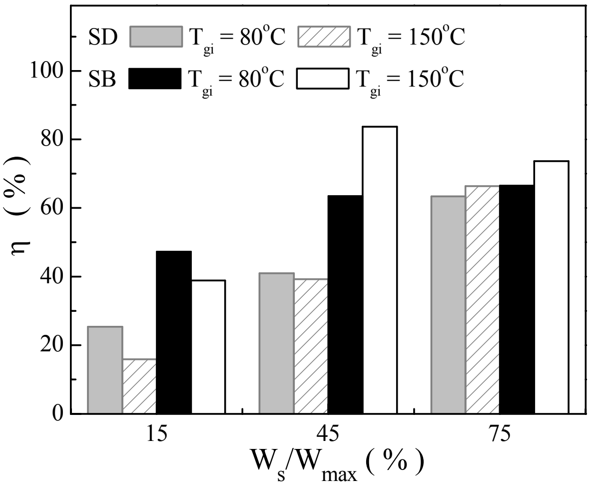


Figure 7: Energetic efficiency ratio as a function of drying system and operating parameters.

CONCLUSIONS

This article presents an investigation of potential of the spray and spouted bed technology for the production of dried extracts of *Rosmarinus officinalis* Linné, an aromatic plant widely employed in the food, cosmetic and pharmaceutical industries. The main approaches adopted in the process development were described and discussed. Special attention was directed to problems of loss or degradation of active compounds during processing.

The powdered *Rosmarinus officinalis* extracts obtained in this work do not have good flow properties and presented high Hausner ratios and compressibility indices, indicating a cohesive or fairly free-flowing tendency and poor compression characteristics. The spray and spouted bed dried extracts had respectively mean loosely packed bulk densities of 320 ± 10 and 430 ± 10 kg/m^3 and tapped bulk densities of 620 ± 30 and of 780 ± 20. The real density, measured with a helium picnometer, was 1493.7 ± 2.9 kg/m^3. The morphological analyses show a product with a large quantity of irregular and polydisperse agglomerated particles with mean diameters varying from 7 to 22 µm.

Higher losses of total phenol (45 to 53%) and flavonoid contents (13.1 to 49.9%) were observed for both the spray and the spouted bed dried extract. In general, the total flavonoid degradation ratio tended to increase with W_s/W_{max} and T_{gi}, whereas the increase in drying gas temperature resulted in a reduction in total phenol concentration.

There were severe performance problems in the spouted bed drying of rosemary extracts, principally excessive product accumulation in the bed and a low product recovery ratio, associated with large high losses of active markers. At present, these drawbacks make the use of this equipment for the production of dried *Rosmarinus officinalis* extracts problematic, and more research is required for their evaluation and solution. Although the performance of spray dryer was better than that of the spouted bed dryer, mainly in terms of product recovery, large losses of active markers were observed.

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NOMENCLATURE

Ac	accumulation ratio	% kg/kg
A	absorbance	UA
C_{pg}	specific heat of the drying gas	J/kg K
C_s	solids content	kg/kg
$\bar{d}_p$	mean powder diameter	μm
D_F	total flavonoid degradation ratio	%
D_P	total phenol degradation ratio	%
Fd	dilution factor	(-)
HR	Hausner ratio	(-)
IC	compressibility index	(-)
Mc	mass collected by the cyclone	kg
M_f	final bed mass	kg
M_i	initial bed mass	kg
Q	spouting gas flow rate	m^3/s
Q_{ms}	gas flow rate at minimum spouting	m^3/s
R	product recovery ratio	% kg/kg
T_E	extractable matter	% kg/kg
T_F	total flavonoid content expressed as quercetin	% kg/kg
$T_{F,ext}$	total flavonoid content in the extractive solution (dry basis)	% kg/kg
T_P	total phenol content expressed as gallic acid	% kg/kg
$T_{P,d}$	total phenol content in the dried extract (dry basis)	% kg/kg
$T_{P,ext}$	total phenol content in the extractive solution (dry basis)	% kg/kg
T_{gi}	inlet gas temperature	°C
T_{go}	outlet gas temperature	°C
U_P	loss on drying	% kg/kg
W_{max}	evaporation capacity of the dryer	kg/s
W_g	mass flow rate of the spouting gas	kg/s
W_s	mass flow rate of the feed composition	kg/s

Greek Symbols

α	significance level	(-)
θ	processing time	s
η	energetic efficiency of the dryer	(-)
λ	latent heat of vaporization	kJ/kg
ρ_b	loosely packed bulk density	kg/m^3
ρ_{bt}	tapped bulk density	kg/m^3
ρ_r	real density	kg/m^3

REFERENCES

Akerele, O., Summary of WHO Guidelines for the Assessment of Herbal Medicines, HerbalGram, 28, 13 (1993).

Al-Sereiti, M.R., Abu-Amer, K.M., Sen, P., Pharmacology of Rosemary (*Rosmarinus officinalis* Linné) and its Therapeutic Potentials, Indian J. Exp. Biol., 37, Nº 2, 124 (1999).

Albu, S., Joyce, E., Paniwnyk, L., Lorimer, J.P., Mason, T.J., Potential for the use of Ultrasound in the Extraction of Antioxidants from *Rosmarinus officinalis* for the Food and Pharmaceutical Industry, Ultrasonic Sonochemistry, 11, 261 (2004).

Andlauer, W., Stumpf, C., Hubert, M., Rings, A., Fürst, P., Influence of Cooking Process on Phenolic Marker Compounds of Vegetables, Int. J. Vitam. Nutr. Res., 73, Nº 2, 152 (2003).

Blanco, M.C.S.G., Ming, L.C., Marques, M.O.M., Bovi, O.A., Drying Temperature Effects in Rosemary Essential Oil Content and Composition, Acta Horticulturae, Nº 569, 95 (2002).

Bott, R.F., Comparative Study of the Production of Dried Extracts of Medicinal Plants by Spray and Spouted Bed Drying, Internal Research Report - Program PIBIC CNPq/USP (2001).

Box, G.E.P., Behnken, D.W., Some New Three Level Designs for the Study of Quantitative Variables, Technometrics, 2, 455 (1960).

Calixto, J.B., Efficacy, Safety, Quality Control, Marketing and Regulatory Guidelines for Herbal Medicines (Phytotherapeutic Agents), Braz. J. Med. Biol. Res., 33, 179 (2000).

Chan, K., Some Aspects of Toxic Contaminants in Herbal Medicines, Chemosphere, 52, 1361 (2003).

Cordeiro, D.S., Produção de Extrato Seco de *Maytenus ilicifolia* Martius ex Reiss pelo Processo leito de Jorro (M.Sc. Thesis), 80 p. (2000).

Cordeiro, D.S., Oliveira, W.P., Technical Aspects of the Production of Dried Extract of *Maytenus ilicifolia* Leaves by Jet Spouted Bed Drying, Int. J. Pharm., 299, N$^{\underline{o}}$ 1&2, 115 (2005).

Brazilian Pharmacopoeia, 3° ed. São Paulo: Andrei, 1988.

Jong, J.A.H., Hoffmann, A.C., Finkers, H.J., Properly Determine Powder Flowability to Maximize Plant Output, Chem. Eng. Prog., 95, N$^{\underline{o}}$ 4, 25 (1999).

Kulevanova, S., Stefova, M., Stafilov, T., Determination of Total Flavonoids and Quercetin in *Hyperici herba* and its Aqueous, Aqueous-ethanolic and Oil Extracts, Acta Pharm., 50, 29 (2000).

Miliauskas, G., Venskutonis, P.R., Van Beek, T.A., Screening of Radical Scavenging Activity of some Medicinal and Aromatic Plant Extracts, Food Chemistry, 85, 231 (2004).

Montgomery, D., Design and Analysis of Industrial Experiments, New York: John Wiley, (1976).

Newall, C.A., Anderson, L.A., Philipson, J.D., Plantas Medicinais – Guia para Profissionais de Saúde, São Paulo: Ed. Premier (2002).

Oliveira, W.P., Bott, R.F., Souza, C.R.F., Manufacture of Standardized Dried Extracts from Medicinal Brazilian Plants, Drying Technology, 24, N$^{\underline{o}}$ 5, 523 (2006).

Prista, L.N., Alves, A.C., Morgado, R., Pharmaceutical Technology, vol II, 4th ed., Fundation Calouste Gulbenkian (1995).

Raghavan, G.S.V., Orsat, V., Electro-technology in Drying and Processing of Biological Materials. Proc. of the 11th Int. Drying Symp., A:73-84 (1998).

Runha, F.P., Development and Analytical Evaluation of Dried Extract of *Maytenus ilicifolia* by Spouted Bed Process, Internal Research Report – Program PIBIC CNPq/USP (2000).

Runha, F.P., Cordeiro, D.S., Pereira, C.A.M., Vilegas, J., Oliveira, W.P., Production of Dry Extracts of Medicinal Brazilian Plants by Spouted Bed Process: Development of the Process and Evaluation of Thermal Degradation during Drying Operation. Trans Ichem., 79 (C), 160 (2001).

Schiavetto, I.A., Caracterização do Extrato Seco de *Rosmarinus officinalis* obtidos pelos Processos Leito de Jorro e Spray Dryer, Research Report - Undergraduate Project, FAPESP, Brazil, 72 p. (2004).

Senna, E.L., Petrovick, P.R., Ortega, G.G., Bassani, V.L., Preparation and Characterization of Spray Dried Powders from *Achyrocline satureioides* (Lam) DC Extracts, Phyt. Res., 11, N$^{\underline{o}}$ 2, 123 (1997).

Souza, C.R.F., Comparative Study of the Production of Dried Extract of *Bauhinia forficata* Link by the Spray Dryer and the Spouted Bed Processes (M.Sc. Thesis), Brazil, 180 p. (2003).

Souza, C.R.F., Oliveira, W.P., Dried Extracts of *Bauhinia forficata* Link Obtained by Spray Drying: Effect of Operating Parameters on Thermal Degradation of Active Compounds and on Physical Properties of the Product, Proc. of the XV Braz. Cong. of Chem. Eng. (CD-Rom) (2004).

Souza, C.R.F., Oliveira, W.P., Comparative Study of the Evaporation Capacity of the Conventional and Jet Spouted Bed Dryers for Liquid Materials, Proc.13th Int. Drying Symp., B, 808, China (2002).

Souza, C.R.F., Oliveira, W.P., Spouted Bed Drying of *Bauhinia forficata* Link Extract: The Effects of Feed Atomizer Position and Operating Conditions on Equipment Performance and Product Properties, Braz. J. Chem. Eng., 22, N$^{\underline{o}}$ 2, 239 (2005).

Souza, K.C.B., Development of Analytical and Technological Methodologies for Obtaining Nebulized Dried Extracts of *Passiflora edulis* Variedade Flavicarpa (M.Sc. Thesis), Brazil, 141 p. (1997).

Teixeira, H.F., Evaluation of the Influence of Pharmaceutical Excipients on Physical, Chemical, Technological and Pharmacological Characteristics of Nebulized Extracts of *Achyrocline satureioides* (Lam.) DC. Compositae - Marcela. (M.Sc. Thesis), Brazil, 146 p. (1996).

WHO, Quality Control Methods for Medicinal Plant Materials, Geneva, Switzerland (1998).