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Study on Optimization Spray Drying Process Conditions for Maximum Stable Granular Aloe vera Gel Powder with a Low Unstable Rate of Aloverose

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ABSTRACT: *In recent years, the use of spray drying for the production of anhydrobiotics has gained the interest of functional food manufacturers, mainly due to cost efficiencies and enhanced product and process flexibility (e.g., enhanced shelf life). In the present work, spray-drying conditions (air inlet temperature and feed flow rate) were optimized for the granulation of the thermo sensitive Aloe vera gel stabilized in a 7.0% arabic gum carrier. A 23 full-factorial experimental design was constructed with air inlet temperature (120, 140, and 160℃) and feed flow rate (6, 7.5, and 9.0 mL/min) as the independent variables and Total Aloverose Contents (TAC), water activity (aw), and cyclone recovery (CR) defined as the dependent variables. The increase in air inlet temperature from 120 to 160℃ induced a significant ($p < 0.001$) reduction in the TAC from 11.8 to 9.4 %. On the other hand, the increase in the feed flow rate from 6 to 7.5 mL/min significantly reduced ($p < 0.001$) the heat-induced viability loss. A further increase in the feeding rate did not further modify the achieved thermo protection, and a detrimental impact of cyclone recovery (reduction) and water activity (increase) of the powder was observed. Using pruned quadratic mathematical models, the optimum spray-drying conditions for the production of maximally stable granulated Aloe vera gel were 133.34℃ and 7.14 mL/min. The physicochemical and structural characteristics of the powders produced were acceptable for application with regards to residual water content, particles mean size, and thermo physical properties to ensure appropriate storage stability under room temperature conditions, with a low unstable rate of Aloverose. Powder particles appeared partially collapse by scanning electron microscope with a spherical shape with surface concavities.*

KEYWORDS *Spray drying process, Optimum production conditions, Maximum stable granulation, Feed rate, Inlet temperature, Aloe vera Gel, Low unstable rate, Aloverose contents.*

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I. INTRODUCTION

Aloes have been used therapeutically, certainly since Roman times and perhaps long before (Morton, 1961, Crosswhite and Crosswhite, 1984), different properties being ascribed to the inner, colorless, leaf gel and to the exudate from the outer layers. During the 12 years since the last major review of *Aloe vera* (L.) Burm.f. gel (Grindlay and Reynolds, 1986) popular interest and use of the gel have increased dramatically. In this country it is now a familiar ingredient in a range of healthcare and cosmetic products widely available and

advertised in shops. The preserved but otherwise untreated gel is also sold as a therapeutic agent in its own right as are various concentrated, diluted and otherwise modified products. This need has been met by a number of wholesalers who get their supplies from plantations in Texas, Florida and Venezuela while new ones are being proposed for Israel, Queensland and East Africa (Jamieson, 1984). This commercial activity has been accompanied by an upsurge of both clinical and chemical research which is reaching more closely towards the active ingredients and their biological activity. There is now less said about doubts as to the efficacy of the material, although there are some warnings of allergic side effects (Klein and Penneys, 1988, Briggs, 1995). Harmful reactions to aloe gel treatment are recorded infrequently (Hunter and Frumkin, 1991, Schmidt and Greenspoon, 1993) but need to be taken seriously. There is still confusion between the leaf exudate and the gel, Morsy and Ovanoviski (1983), Natow (1986) and Duke (1985) where a great number of folk medicine uses are described and Ahmad et al. (1993). However, many commentators clearly distinguish between the two parts (Watson, 1983, McKeown, 1987, Capasso et al., 1998) and describe in some detail how the gel is prepared (McAnalley, 1988, McAnalley, 1990, Agarwala, 1997). At one time there was much discussion about the relative efficiency of 'decolorized' and 'colorized', i.e. with exudate components, gels (Danof, 1987, Agarwala, 1997). There is also a feeling that some of the variable results reported in the literature may be due to treatment of the gel subsequent to harvest (Fox, 1990, Marshall, 1990, Briggs, 1995, Agarwala, 1997).

A number of reviews have appeared in recent years covering various aspects of aloe gel use, as well as much commercial literature. Exaggerated claims are still being made and although doubts as to the substance's efficacy are more muted, there is still room for the caution which has been voiced (Hecht, 1981, Marshall, 1990). The emphasis is changing towards definition of the active constituent or constituents so that they can be used accurately in formulations (Reynolds, 1998).

The term aloverose refers to the marker component of aloe gel, which, when administered in sufficient amounts (typically greater than 10% of a product), provides beneficial health effects to the host.[23] Incorporating aloverose into real-world food systems can be challenging for food manufacturers, not only because of the differences in aloe's growing environment, but also because it is sensitive to the harsh processing and chemical conditions used in the food processing industry. These conditions can cause aloverose damage through osmotic and thermal stress (e.g., heat processing, high solute concentration) and increased redox potential. [24–26] Additionally, suboptimal storage conditions can result in changes in physical state (e.g., aging and spoilage), which can induce physical and biochemical reactions that can negatively affect aloverose content. [24–26] In the past few years, the aloe industry has experienced a significant increase in market share due to the introduction of a wide range of products containing aloe, including traditional foods, cosmetics and pharmaceuticals, and health foods for direct consumption.[25,27-30] Dehydration processes such as freeze drying, spray drying, freeze-spray drying, vacuum, and fluidized bed drying are among the most common practices to produce anhydrous products for incorporation into food systems while maintaining the aloverose content after processing.[24] Freeze drying is considered one of the best practices for reducing heat damage, but spray drying is more advantageous in terms of cost, energy, and throughput.[31]

Although there is a wide demand for inexpensive spray-dried aloe powder, the use of spray drying as a standard production technique is challenging due to the negative impact on the aloverose content during drying (if not properly optimized) and the decreased stability of the spray-dried aloe gel during product production. [24,25,31] It has been demonstrated that the aloverose content due to heat is mainly related to the change in the physical state of the aloe gel. For example, there may be phase transition from crystalline to gel state, modification of aloe gel rheology, peroxidation of aloeverose, structural modification of macromolecular structure (e.g., unfolding of glycoproteins or minerals), and biochemical changes of other polysaccharides in the aloe gel.[24,32] In addition to carrier-aloe gel interactions, the influence of the spray drying process is related to process parameters (inlet and outlet air temperature, feed flow rate, residence time in the drying chamber, design parameters of the drying chamber, temperature of the drying medium, etc.), thermodynamic processes (heat and mass transfer rates during droplet dehydration), drying kinetics (effects of steady and reduced drying rates), and biology of the aloe to be powdered (cultivar and type, growth period and condition, acclimatization to thermal or osmotic stress conditions).[25,33–37] Gardiner et al.[3] reported that the content of aloeverose decreased with increasing outlet temperature. The same researchers reported that the aloeverose content was optimized when using an air outlet temperature of 70–80 C. Using a higher feed rate also reduced heat-induced cell damage, but this approach resulted in increased water activity of the final product and altered morphological characteristics of the spray-dried powder. [38,39]

The effect of carrier materials on aloeverose content via spray drying has been previously studied.[27-29,33-37] The presence of components that can induce significant decreases in the total solids concentration and melting point of the carrier fraction and the fine particles have been reported to significantly affect the structural integrity of aloe gels and control the osmotic pressure leading to aloe gel rupture.[24] For this reason, materials that ensure excellent particle size distribution and impart acceptable powder functional properties (porosity, free-flowing ability, anti-clumping, and good wetting and dispersing properties) are often selected as carrier

materials, as well as other components that exhibit thermal protection functions such as disaccharides (lactose, sucrose, or trehalose), dextrose, or polyols (mannitol, sorbitol) or act as other polysaccharides (fructose and oligosaccharides) in aloe gels. [24,25,33-36,38]

Recently, Shabnam et al. [27] demonstrated that the use of complex carbohydrate-protein systems as potential carriers for anhydrous products can improve their performance during storage. In this study, the effects of spray drying conditions (inlet temperature and feed rate) on microfiltered heat-sensitive aloe gels in carrier systems consisting of Arabic gum and Xanthan gum systems were investigated. The optimal spray drying conditions required to produce a highly stable aloe gel dry powder formulation with acceptable physicochemical and structural properties were determined.

II. RESEARCH METHODS

A. Preparation of Aloe Gel

3 kg of *Aloe vera* (KimJungMoon Aloe, Jeju, Korea) leaves grown for more than 3 years were collected and the surfaces of the leaves were washed clean with water. The upper and lower parts of the washed aloe leaves were cut into a certain size using a conventional method. The cut aloe leaves were placed in a water bath containing 500 ml of distilled water, set to room temperature (20℃), and then immersed for 30 minutes. The outer skin of the immersed aloe leaves was removed and crushed twice with a juicer to produce aloe gel.

B. Preparation of the Drying Media

5% Arabic gum (C Dry MD 01910, Cargill Ltd., Manchester, UK) and 0.1% Xanthan gum (Fisher Chemicals, Loughborough, UK) were mixed together and balanced with distilled water to 100 g. The solution was allowed to fully hydrate for 1 h at room temperature under magnetic stirring and then heat treated at 90 C for 10 min to destroy pathogens and allow complete protein denaturation. The Arabic gum-Xanthan gum aliquots were rapidly cooled to room temperature using an ice bath and the aloe gel was suspended.

C. Spray Drying and Storage of Aloe Gel

The prepared aloe gel was dried using a Buchi B-290 laboratory spray dryer (Buchi, Flawil, Switzerland), and the carrier aliquots were kept under low-speed agitation using a magnetic stirrer throughout the spray drying process. A X_{12} 2^3-factorial design of experiments was used to establish the optimal spray drying conditions (inlet and feed rates) in terms of maximum aloeverose content retention and cyclone recovery and minimum moisture level (Table 1). The spray dryer was operated at three different air inlet temperatures (120, 140 and 160℃), and the feed flow rates (6, 7.5 and 9) mL min^{-1}, smf drying air flow rate (35 m^3 h^{-1}) and compressor air pressure (0.5 MPa) were kept constant throughout the drying process. The outlet temperature varied proportionally with the air inlet temperature and feed flow rate conditions (Table 2). The dried probiotic formulation was collected from the cyclone separator vessel, placed into sealed glass vials, and stored at room temperature in a desiccator containing saturated lithium chloride (LiCl, Fisher Scientific) solution to provide dry conditions (a_w = 0.11).

Table 1. Codification of the independent variables (air inlet temperature and feed flow rate) used for the construction of the response surface design

Independent variables (factors)	Factor levels		
	-1	0	+1
X_1 = Inlet air temperature (℃)	120	140	160
X_2 = Feed flow rate (mL=min)	6.0	7.5	9.0

D. Design of the Experiments

A full-factorial design (n = 32) was used for this course of experiments with air inlet temperature (X_1) and feed flow rate (X_2) as factors and the total viable counts (TVC) after spray drying, the water activity of the spray-dried product (a_w), and the cyclone recovery percentage as the responses. Full-factorial design of experiments (DOE) have previously been successfully applied in a wide range of applications; for example, quality and process optimizations studies in food systems and consumer preference studies. [28–30]

Although full-factorial DOE approaches are generally regarded as time and cost consuming due to the large number of experiments required, they do offer the best practice approach for process or product optimization where the factor interactions cannot be neglected. [40]

Analysis of variance (ANOVA) was performed to estimate the significance ($p < 0.05$) of the main effects (linear and quadratic) and their interactions (linear, linear-quadratic, and quadratic–quadratic). The effects of the inlet air temperature and feed flow rate were modeled in a full quadratic mathematical model using response surface methodology as described in Eq. (1):

$$Y = \beta + \alpha_1 X_1 + \alpha_2 X_1 + \alpha_{11} X^2_1 + \alpha_{22} X^2_2 + b X_1 X_2 + c_1 X^2_1 X_2 + c_2 X_1 X^2_2 + {}_d X^2_1 X^2_2 \quad (1)$$

where Y denotes the response variable, β is the intercept constant, a_1 and a_2 are the main linear effects, a_{11} and a_{22} are the main quadratic effects, and b, c, and d are the linear–linear, linear–quadratic, and quadratic–quadratic interaction coefficients, respectively. The significance of the parameters was tested using t-test. ANOVA was also used to evaluate the performance of the generated models, through the separation of residual variation into lack of fit and pure error linked to the replicate error at the central point. Goodness of fit (R^2) was calculated for each model as an estimation of the upper bound of the predictability of the model. In addition, the goodness of prediction (Q^2) was calculated as a measure of the predictive power of the model as described by Eq. (2):

$$Q^2 = \text{SS} - \text{PRESS} / \text{SS} \quad (2)$$

where SS and PRESS denote the sum of squares of the response values and the prediction of residual sum of squares, respectively. [41]

Table 2. Effect of spray drying conditions (air inlet temperature and feed flow rate) on discharge temperature, water activity, aloverose content of aloe gel and powder recovery in cyclone separator

Treatment (%)	Inlet temperature(℃)	Feed rate (mL/min)	Outlet temperature(℃)	Water activity	Aloverose Content(log/g)	Cyclone Recovery(%)
KJM F/D	120	6	66.5 ± 0.5	0.175 ± 0.03	9.00 ± 0.18	66.6 ± 0.5
GA 5	120	7.5	63.1 ± 0.9	0.199 ± 0.02	8.91 ± 0.12	63.2 ± 0.8
GA 7	120	9	60.0 ± 1.4	0.243 ± 0.11	8.87 ± 0.01	60.1 ± 1.7
K-GA 7	140	6	82.7 ± 1.7	0.138 ± 0.16	8.65 ± 0.12	70.7 ± 1.5
GA 10	140	7.5	76.9 ± 1.2	0.154 ± 0.02	8.55 ± 0.22	66.8 ± 1.6
GX 0.15+GA 5	140	9	72.0 ± 1.9	0.197 ± 0.01	8.41 ± 0.16	62.8 ± 1.9
GX 0.15+GA 7	160	6	91.5 ± 0.4	0.112 ± 0.04	7.37 ± 0.11	73.9 ± 0.7
GA	160	7.5	88.5 ± 0.2	0.132 ± 0.01	7.81 ± 0.08	70.2 ± 0.0
GX	160	9	85.1 ± 1.0	0.159 ± 0.02	8.19 ± 0.20	67.6 ± 0.0

E. Analytical Method for Aloeverose Content in Aloe Gel Spry Dried Powders

This study intended to standardize the method for aloeverose (total polysaccharide), which is a functional marker for aloe gel in Korea. We used 9 spry dried powders and commercial aloe gel products, certified as Health Functional Food by Korea Food and Drug Administration (KFDA), including powder, solution, jelly, tablet and capsule, to optimize the analytical condition of dialysis and phenol-sulfuric acid reaction in aloeverose analysis. The optimal conditions for aloeverose analysis included 1 L water for dialysis and change 3 times for 24hr against 25 mL prepared sample solution. Validation test showed lower than 5% of coefficient of variation (CV) in intra-, intraday validation in spry dried powders and 9 types of commercial products. In inter-person and inter-laboratory validation with 3 persons from 3 different laboratories, CV (%) were 5.50 and 6.64 respectively. The linearity of aloeverose analysis was assessed using 9 serial concentrations of spry dried powders.

The aloeverose content after the spray drying process was calculated according to the following formula:

$$\text{Aloeverose content (mg/g)} = N / N_0 \quad (3)$$

where N_0, N represent the aloe vera content before and after the spray drying process. [33-37]

F. Characterization of the Optimized Final Product

1) Moisture Content and Water Activity

The moisture content was calculated according to American Association of Cereal Chemists (St. Paul, MN, USA) method AA-15A (Approved Methods of Analysis, 11th ed.). Two grams of the powder were placed in aluminum pans and dried at 105 ℃ for 24 h. Residual moisture content was calculated according to the formula

$$\% \text{ moisture} = 100 \times w_f - w_i / w_i \quad (4)$$

where w_i and w_f are the weights of the dry aloe gel formulations prior to and after dehydration at 105 ℃. Water activity was measured using an Aqua Lab water activity meter (Aqua Lab, 3TE, Decagon, Pullman, WA, USA).

2) DSC Measurements

A standard power-compensated Perkin Elmer DSC-7 (Perkin Elmer Ltd., Beaconsfield, UK) was used for calculation of the glass transition temperature of the optimized formulation. A small portion (15–20 mg) of the powder was weighted in a high-pressure, stainless-steel pan and heated from -30 to 150 ℃ at a rate of 10 ℃/min. A double heating–cooling scanning step was performed, and thermal properties (onset, midpoint, and offset glass transition temperatures and specific heat capacity change, ΔC_p) were calculated using Mettler Toledo Star (Columbus, OH, USA) software from the second heating step thermographs.

3) Particle Mean Size Analysis

The particle mean size analysis was performed on a laser diffraction particle size analyzer equipped with the Tornado dry powder system (LS 13320, Beckman Coulter, USA). The Fraunhofer theory was used for the determination of the mean diameters of the microparticles. The volume distributions of the samples were calculated and the results are presented as mean particle size diameter.

4) Color Measurement

One gram of powder was put in plastic cuvettes and color measurements were performed using a Hunter lab (Color Quest XE, HunterLab, Reston, VA, USA) colorimeter. The CIE Lab color scale was used to measure the L^* (black to white), a^* (red to green), and b^* (yellow to blue) parameters. The total color difference, ΔE^*, between a white standard tile (L^* = 92.59, a^* = -0.78, b^* = 0.67) and each individual dry aloe gel formulation were calculated according to the formula

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (5)$$

where ΔL^*, Δa^*, Δb^* are the luminosity, redness, and yellowness intensity difference from the control sample.[19]

5) Hygroscopicity

The hygroscopicity of the aloe gel powders was determined according to the procedure described by Iris Schmitz-Schug *et al*. [35] More specifically, a 1 g sample of the powder was placed in a desiccator equilibrated at 75% relative humidity containing a saturated sodium chloride (NaCl) solution at room temperature. Samples were kept for 7 days and hygroscopicity was calculated gravimetrically according to the formula

$$\text{Hygroscopicity (g of } H_2O \text{ per g of product)} = 100 \times m_f - m_i / m_i \quad (6)$$

where m_i and m_f express the moisture of the samples prior to and after storage at 75% relative humidity.

6) Dissolution

The dissolution capacity of the powders was calculated according to the method of Iris Schmitz-Schug *et al*. [35] One gram of powder was added to 50 mL of distilled water and dispersed under magnetic stirring (Ika GmbH, Germany) at 892 rpm using a 2 mm × 7 mm stirring bar. The time required for complete dissolution of the powder was recorded.

6) Morphological Characterization

For visualization of the morphology of the microparticles, a small amount of powder was carefully deposited onto carbon tabs (Agar Scientific, Stansted, UK) and coated with carbon (agar turbo carbon coater) to improve conductivity. Scanning electron microscope analysis (SEM) was performed on an FEI Quanta 3D 200 dual-beam focused ion beam scanning electron microscope (FIB-SEM). The images were acquired using secondary electron imaging at an accelerating voltage of 5–15 kV.

III. RESULTS AND DISCUSSION

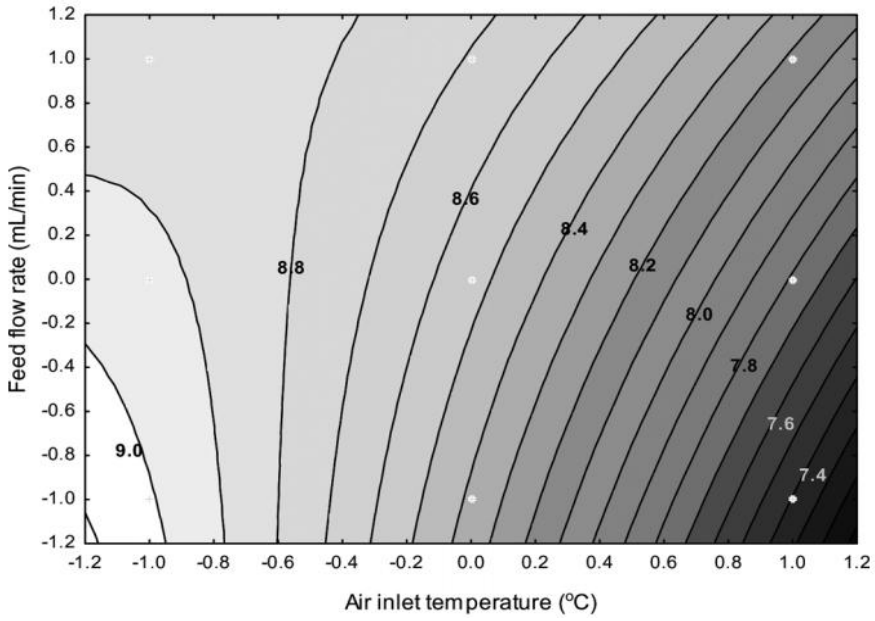
A. Aloeverose content throughout the Spray-Drying Process

The impact of the tested spray-drying conditions (inlet air temperature and feed flow rate) on the content of aloeverose throughout the process is displayed in Table 2 and the resultant model is graphically illustrated in Fig. 1.

As can be seen from Fig. 1, a reduction in the air inlet temperature was in almost all cases linked to an enhancement in aloeverose content and at mid and high air inlet temperatures an elevation of the feed flow rate increased aloeverose content, although this relationship was reversed at low air inlet temperatures. The most significant change was observed when the outlet temperature was reduced from 91.5 to 60 C; over this temperature range the content of aloeverose increased from 2.5 to 84%. It is well established that the loss of aloeverose content during convective thermal processing is related to aloe gel injuries resulting from the combined effect of heat and mechanical stress. Examples include the denaturation of the informational macromolecules (total polysaccharides), damage to aloe gel, dehydration of cytoplasmic membranes, lipid peroxidation, and rupture and collapse of cell membrane due to water removal. [24,32,34,42,43]

Under excessive droplet heat transfer rate conditions (increase of the T_d - T_g driving force), the integrity of the cellular membranes can be lost due to crystalline to rubbery state transitions, which are responsible for the increase the membranes fluidity, leading eventually to the cells' fate.[24] The changes observed as a result of alterations in the flow rate are a result of changes in the heat and mass transfer kinetics at the air–solid interface. Generally, the elevation of the feed flow rate causes a reduction in the droplets' surface temperature, which causes changes in both heat and water diffusivity,[44] consequently reducing the physical damage to the cell membranes. As can be seen in the results of the ANOVA (Table 3), both linear and quadratic coefficients are significant for air inlet temperature and the linear coefficients are significant for feed flow rates (p<0.001).

Fig. 1. Effects of air inlet temperature and feed flow rate on the suitable contents of aloeverose after spray drying



B. Effects of Processing Conditions on Water Activity and Cyclone Recovery

The water activity of the spray-dried powder was significantly (p<0.001) affected by air inlet temperature and feed flow rate as displayed in Fig. 2. In general, inlet air temperature was the more impactful factor when compared to feed flow rate for controlling water activity and residual water content (data not shown) of the finished powders; this can be seen in Fig. 2 and is further detailed in Table 3. Products produced with the highest air temperature and lowest feed flow rate resulted in the driest formulations. Low aw values and residual moisture contents (<4-5% w/w) are prerequisites for the commercial production of spray-dried powders with good handling characteristics such as high flow ability, low stickiness and agglomeration, as well as maximum aloeverose content.[44] The residual water content ranged from 1.7 to 5.4% w/w (data not shown), which complies with standard acceptable moisture levels for spray-dried powders.[45] At conditions of low water activity, the matrix moves from the rubbery state toward the glassy state and, thus, water mobility is reduced. This inhibits cell metabolic activity of the aloe gel, leading to extended shelf life. [25,28,36,45]

Both feed rate and inlet temperature significantly impacted cyclone recovery. In both cases this could be described by a linear relationship. In general, spray-drying yield was maximized when the spray dryer was operated at high air inlet temperatures and low feed flow rates (Fig. 3). The amount of the dried product recovered via cyclone separation is influenced by many engineering and product parameters, such as drying air flow and local velocities; the spatial geometry of the separator; and the adhesiveness and cohesiveness of the

particles while interacting with the drying chamber.[45,46] In our work, the air flow was kept constant, and thereby the parameters that affect the surface stickiness of the micro particles such as the hygroscopicity, glass transition temperature, moisture and thermal diffusivity of the carrier material, temperature of the droplets obtained in the drying chamber, etc., probably control the achieved powder recovery at the cyclone separator. [45] According to the findings of Khaled Almansour et al.,[45] the presence of ingredients that act as plasticizers; for example, sugars increase the surface stickiness of the dried particles due to the increase in the glass transition–surface temperature gradient. Thus, as the T_d - T_g value increases the droplets move toward the rubbery state, sticking on the drying chamber surface and reducing the powder recovery rates. The effect is dependent on both heat and mass diffusion rates, because water acts as a plasticizer for macromolecules.[47] In our case, the combination of high air inlet temperature and low feed flow rates led to less particle stickiness probably due to the lower residual contents achieved by the sufficient heat penetration and core-to-droplet surface water diffusion rate.

Table 3. Regression coefficients and their significance levels for pruned mathematical models used for the prediction of total suitable contents of aloeverose, water activity, and powder recovery at the cyclone separator (ns = not significant)

	Suitable aloeverose contents		a_w		Cyclone recovery	
	Coefficient	*p*-Value	Coefficient	*p*-Value	Coefficient	*p*-Value
Intercept	8.417	<0.001	1.57E-01	<0.001	67.9	<0.001
X_1	- 0.569	<0.001	-3.57E-02	<0.001	3.633	<0.001
X_2	0.089	<0.001	7.00E-03	0.018	-3.350	<0.001
X^2_1	0.156	0.008	2.90E-02	<0.001	ns	0.895
X^2_2	ns	0.884	9.00E-03	0.003	ns	0.878
$X_1 X_2$	0.238	<0.001	-5.25E-03	0.013	ns	0.872
$X^2_1 X_2$	ns	0.705	ns	0.352	ns	0.711
$X_1 X^2_2$	ns	0.478	ns	0.828	ns	0.408
$X^2_1 X^2_2$	ns	0.897	ns	0.267	ns	0.802
SS	7.539		0.039147		461.4	
Pure error	0.389		4.62E-04		20.27	
Lack of fit (*p*-value)	0.0154 (0.946)		0.001216 (0.112)		1.535 (0.962)	
R^2	0.946		0.976		0.952	
R^2_{adj}	0.936		0.971		0.949	
PRESS	4.05E-01		9.37E-04		21.8	
Q^2	0.943		0.973		0.953	

Fig. 2. Effects of air inlet temperature and feed flow rate on the water activity of the spray-dried powders

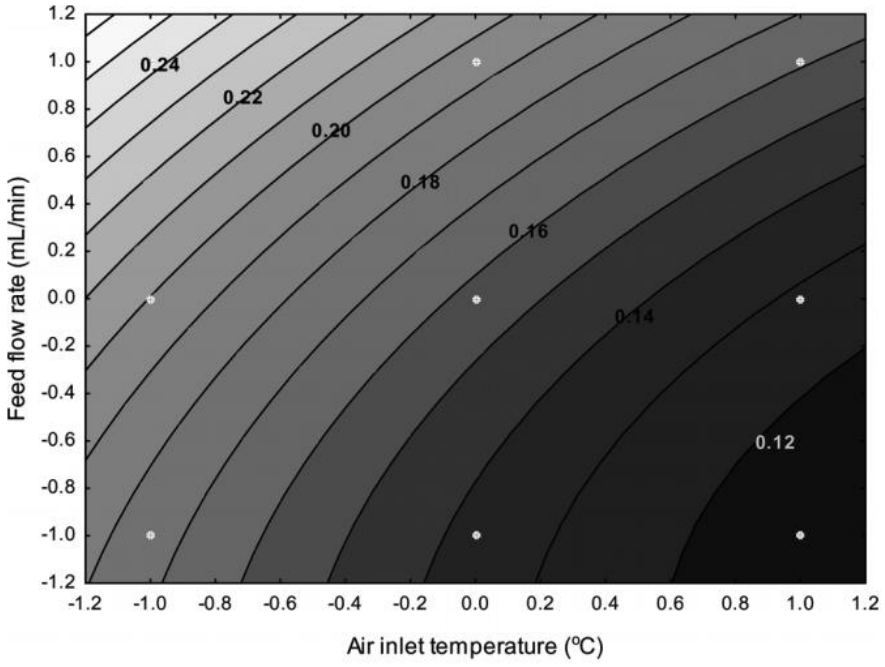
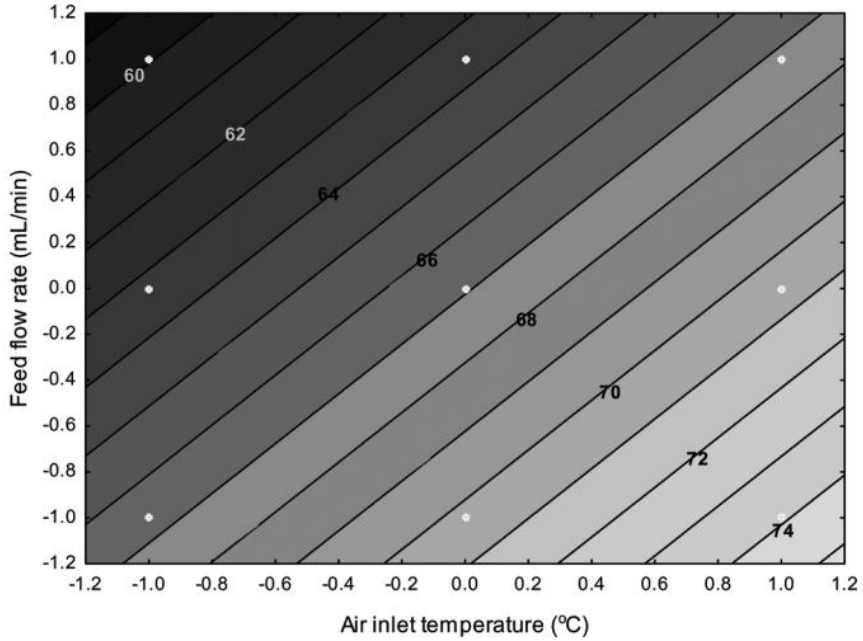


Fig. 3. Effects of air inlet temperature and feed flow rate on the powder recovery at the cyclone separator



C. Optimization of Processing Conditions and Model Verification

It is not possible to simultaneously maximize the content of aloverose while minimizing water activity and maximizing cyclone yield. Therefore, compromises must be made and an optimal compromise must be sought. Optimal operating conditions that produced both suitable aloverose particles and a stable dry powder were calculated using a desirability function,[34] which is further detailed in Eq. (7):

$$\text{Desirability} = (\text{TVC} \times a_w \times \text{CR})^{1/3} \quad (7)$$

where TVC is the total aloverose content, a_w is the water activity, and CR is the powder recovery in the cyclone separator. The desirability targets were set at 1 (maximum) for TVC and CRA and 0 (minimum) for water activity. As can be seen in Table 4, desirability was maximized when X_1 = -0.333 and X_2 = -0.233, which corresponds to an air inlet temperature of 133.54 ℃ and a feed flow rate of 7.14 mL = min. In order to validate the constructed mathematical models, a spray-drying experiment using the same carrier system and operating the spray dryer as close as possible to the optimum conditions (134 ℃ and 7.2 mL = min) was carried out. The error between the predicted and observed values was 2.3% for a_w, 0.35% for TVC, and 1.2% for CR, verifying the predictive powder of the constructed models. Statistically, the model was further justified by the lack of fit (p > 0.05) and Q^2 values shown in Table 3.

Table 4. Optimum spray-drying conditions and validation of the pruned mathematical models constructed for the prediction of total aloeverose contents, water activity, and powder recovery at the cyclone separator

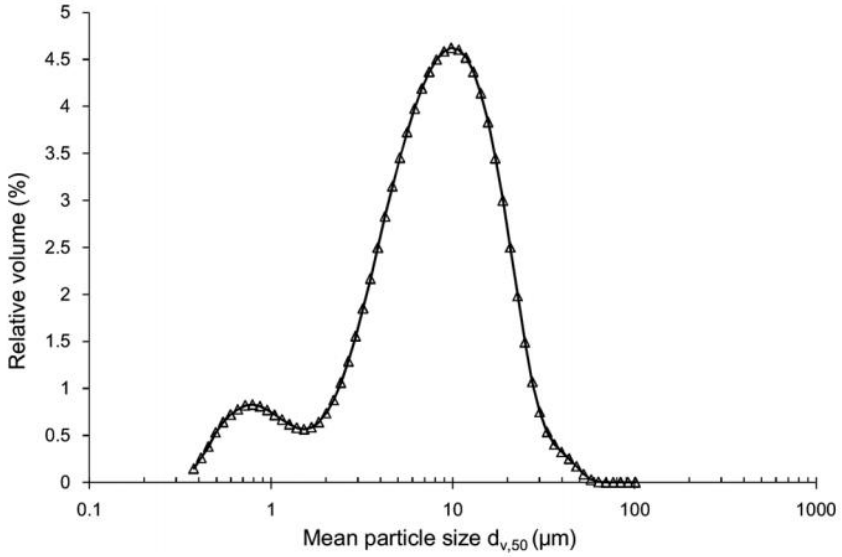
Factor		Optimized conditions		Operating conditions	
Inlet temperature (℃)		133.54		134	
Feeding rate (mL=min)		7.16		7.2	
Outlet temperature (℃)		73.19		73-74	
Responses					
Water activity		Total aloverose contents (log/g)		Cyclone recovery (%)	
$a_{w[\text{observed}]}$	$a_{w[\text{predicted}]}$	$\text{TVC}_{[\text{observed}]}$	$\text{TVC}_{[\text{predicted}]}$	$\text{CR}_{[\text{observed}]}$	$\text{CR}_{[\text{predicted}]}$
0.171	0.174	8.59	8.62	68.30	67.47

D. Characterization of the Physicochemical and Structural Characteristics of Optimized Microparticles

The physicochemical and structural properties of the microparticles produced under the optimized spray dryer operating conditions are detailed in Table 5. Particle size analysis revealed a bimodal mean size distribution (Fig. 4) that was characteristic of spray-dried powders with high bulk (tap) density; in general, bimodal distributions pack most efficiently as the smaller particles are included in the voids between the larger microparticles.[48] The volume-weighted mean diameter ($d_{V,50}$) of the microparticles was 10.96 mm, which is

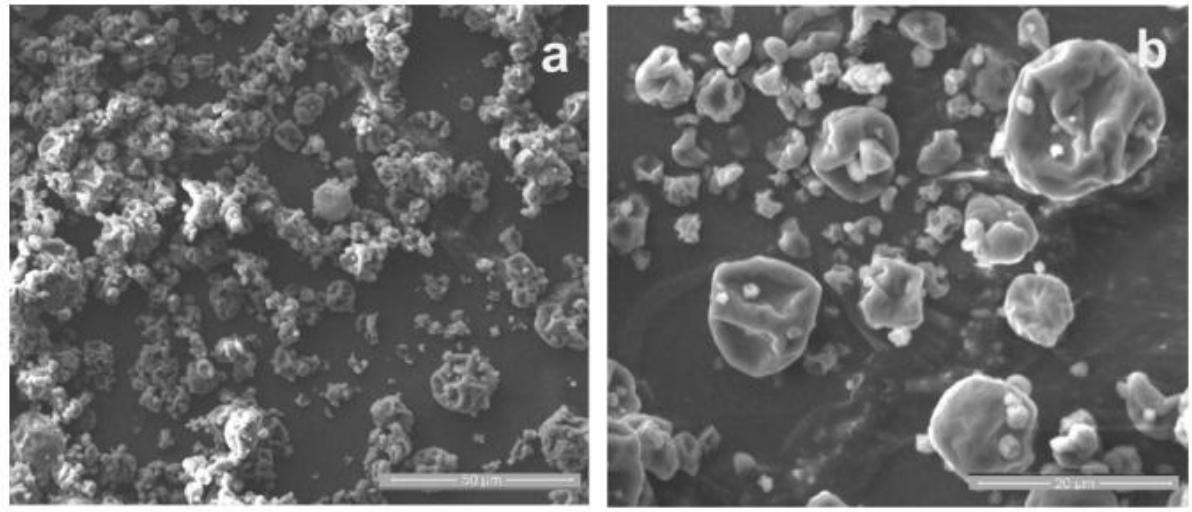
comparable to the values (10–20 μm) reported in the case of other spray-dried aloe gel formulations.[33-37,49] The hygroscopic character of the powders (0.174 g/g of absorbed water) can be explained by the presence of arabic or xanthan gums (both of which have hygroscopic properties), although the hygroscopicity values were much lower compared to other aloe gel dry formulations.[34-36] The glass transition temperature of the powder was 59 ℃, suggesting that the matrices can be stored under chilling or room temperature conditions while retaining their glassy state, irrespective of the plasticizing effect of arabic or xanthan gums, and free water in the products. [47] The spray-dried powders had a lower luminosity (L^*) and higher yellow color intensity compared to other aloe gel-based spray-dried formulations,[34-36] which was probably due to the presence of the arabic or xanthan gums. Similar results have been also reported by other researchers for the color difference (ΔE^*) for the white standard, which was 2.87; this color threshold is normally quoted for perceivable differences ($\Delta E^* = 3$).[50]

Fig. 4. Particle mean size distribution of the optimized powders containing aloeverose



The morphology of the microparticles is illustrated in the SEM micrographs shown in Fig. 5. The particulate structure of the product generated under the optimum spray-drying conditions had a partially collapsed structure, which is characteristic of many spray-dried powders analyzed under vacuum, that can be described as a deflated, flat, ball-like, spherical particles. The heat transfer rate and the water diffusion rate from the surface to the core of the droplets as well as the presence of arabic or xanthan gums ingredients critically affected the microstructure of spray-dried matrices, with intermediate heat and mass transfer rates (intermediate air inlet temperature and feed flow rate) to induce the highest collapse of the particles.[51]

Fig. 5. SEM micrographs of the microparticules containing aloeverose produced under the optimized spray-drying conditions: (a) scale bar 50 mm and (b) scale bar 20 mm



The inactivation rates of the powders stored for 30 days at chilled temperatures (4 ℃) and ambient room storage conditions (25 ℃) in sealed, airtight vials were calculated by fitting the TVC data to a first-order reaction model, which has successfully has been used in previous studies. [27,34-36,50] The total sutable contents of aloeverose were 8.55 and 7.48 log/g after 30 days of storage at 4 and 25 ℃, respectively, indicating a good storage stability of the aloe gel powders.

IV. CONCLUSION

In the present work, we showed that optimization of the spray-drying process is essential for the production of dry spray powders containing viable thermos sensitive aloe gel and aloverose. The use of intermediate air inlet temperatures and feed flow rates are required to provide sufficient survivability of aloe gel and aloverose through spray drying while retaining good powder recovery rates at the cyclone separator and low residual water activities. In addition, the microparticles produced at the optimized spray-dried conditions were characterized as having acceptable physicochemical properties (total color, glass transition temperature, hygroscopicity). Although the results are promising and show that through compromises in techno-functional properties, yield, and shelf life, high-quality anhidrotic powders can be produced, it is anticipated that the shelf life of powders and the content rates of aloe gel can be further improved by contintinual development of the carrier system and thermal profile of the drying chamber (through chamber geometry optimization and dynamic regulation of exhaust air).

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