Drying by desorption: A tool to determine spray drying parameters

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ABSTRACT

The most frequently used technique for dehydration of dairy products is spray drying. This is an effective method for preserving biological products as it does not involve severe heat treatment and it allows storage of powders at an ambient temperature. Due to the variety and complexity of the concentrates to be dried, a more rigorous understanding of spray drying based on physico-chemical and thermodynamic properties has now become necessary. However, the current state of the art and knowledge do not allow determination of the parameters of spray drying of dairy products. The only way to determine these parameters is to perform several complex and expensive experiments with spray dryer pilots. In this study, a new method was developed to evaluate the ratio of bound to unbound water by using drying by desorption. The results, combined with thermodynamic and physico-chemical parameters (such as, absolute and relative humidity of air, total solids and temperature of concentrate, and air flow rate), provide more precise determination of certain spray drying parameters such as inlet air temperature and mass flow rate. More than 50 experiments were performed to correlate calculated and measured parameters in a pilot plant (Bionov) using water, skim milk, infant formula milk, caseinate maltodextrin and other food concentrates. The results showed that the difference between the calculated and measured inlet air temperature was less than 5%, the determination coefficient being close to 0.96.

The economic value of this system is obvious, because it is easy to anticipate the spray drying parameters by using a controller integrating the water availability of the concentrate and certain thermodynamic parameters. Software based on this step was developed (SD²P®, spray drying parameter simulation and determination) and registered at the APP (Association pour la Protection des Programmes).

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1. Introduction

Drying (consisting of lowering water activity by water elimination) is an effective method for preserving biological products, since it does not involve severe heat treatment and it allows storage at an ambient temperature. Large amounts of liquid dairy products (skim and whole milk, whey, various fractions resulting from membrane filtration and chromatographic separation) and food are dried in order to produce feeds, food and ingredients. Most of these powders are spray dried. This process consists in spraying the concentrated liquid in droplets of about 50 μm into a large drying chamber containing air heated at around 200 °C. The temperature of the product itself lies between the wet bulb temperature and the temperature of the outlet air, i.e. it remains below 100 °C. Since drying occurs within a few seconds, thermal damage

is limited. Classical spray dryers are combined with a fluid bed in the wet zone, which usually agglomerates the fine powder coming from the drying chamber, completes the drying process and cools the powder. In recent three-stage installations, another fluid bed with agglomeration and additional drying functions is included at the bottom of the drying chamber (Sougnez, 1983; Masters, 2002).

There have been few scientific or technical studies on the powder quality obtained from spray drying related to the process parameters, physico-chemical composition or microbiology of the concentrates. Manufacturers have acquired expertise in milk and whey drying processes through an empirical approach. However, due to the variety and complexity of the mixes to be dried, more rigorous methods based on physico-chemical and thermodynamic properties have now become necessary.

Droplet size distribution, droplet trajectories, air flow patterns, and internal and external mass and heat transfer during drying show complex dependence on flow rates (air and concentrate), and physical characteristics depend on the product, the properties

of the material to be dried and the design of the atomizing device and the drying chamber (Ferrari et al., 1989; Masters, 2002). Pisecky (1981) studied the parameters controlling powder moisture content and the influence of various factors on bulk density in a spray drying operation. He showed the different properties of dairy powders which are influenced by processing. Because of this complexity, it is difficult to devise a reliable mathematical model to determine the parameters and performance of a spray dryer as a function of the design variables, operating conditions and feed properties (Ferrari et al., 1989). Several authors have studied the moisture transfer mechanisms for a liquid food droplet during drying (Furuta, 1992), the drying kinetics for a single droplet of skim milk (Kerkhof and Schoeber, 1974; Ferrari et al., 1989) and a comprehensive model based on control of milk drying processes (Chen, 1994). These studies have demonstrated major unreliability for various reasons in these models of drying processes (Ferrari et al., 1989; Furuta, 1992; Chen, 1994), including the prediction

of the mean droplet size and the dimensions of the atomizer used, hydrodynamic interaction of air and sprayed liquid, agglomeration of droplets after atomization, history of shrinkage; desorption isotherms, internal air flow profiles, and drying conditions (air and concentrate flow rate, absolute and relative humidity, etc.).

Improvement in the quality of new dairy and food powders can be obtained only with the integration of new parameters such as glass transition temperature, water activity (Schuck et al., 2007), residence time distribution (Jeantet et al., 2008), surface composition and surface temperature (versus drying time). The latter consists of drying modeling based on heat and mass transfer and balance equations. Two approaches can be considered: an overall approach, developed by Schuck et al. (2005), in which the model results in a mass and energy balance over the entire dryer, which can be a black box between both ends that which efficiently predicts the settings and overall performance of the spray dryer before production starts for varying weather/product conditions, and a micro approach developed by Chen and Lin (2005), Patel et al. (2005a,b) and Lin and Chen (2005, 2006, 2007) which considers the drying kinetics and balance at the droplet level to determine information regarding the condition of the droplet (temperature/water content) as it dries. This reaction engineering approach makes it possible to model the drying droplet of various dairy products and is a new and interesting approach for improved control of the surface temperature of the droplet and subsequently the powder. This model can be used to predict certain aspects of powder quality/functionality. Whatever the approach used, one difficulty remains: i.e. how to take into account the water availability in the product, except for pure water.

The aim of this study was to propose a new method of drying by desorption in order to determine major drying parameters according to food components in relation to their interactions with water (bound and free water) and linked to water transfer kinetics.

2. Materials and methods

2.1. Drying by desorption

Concentrates were dried at 45 °C (constant temperature) in a device (Rotronic, Bassersdorf, Switzerland) which consists of a two compartment cell, one (the smallest) containing the product
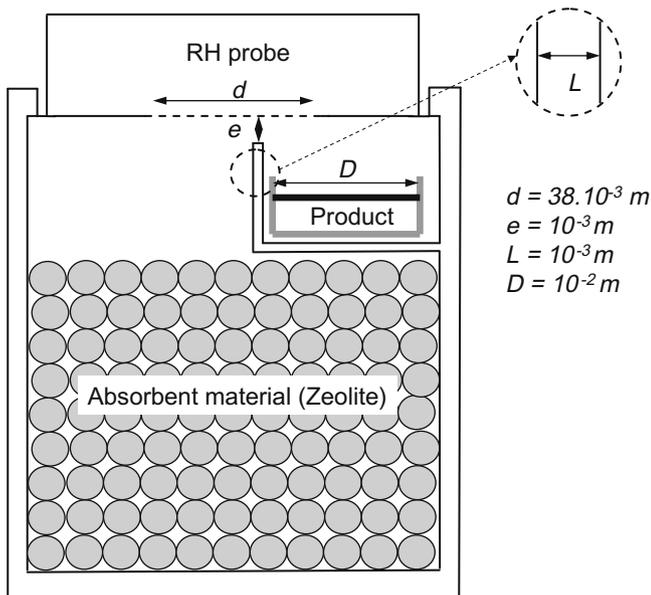

$d = 38.10^{-3}$ m
$e = 10^{-3}$ m
$L = 10^{-3}$ m
$D = 10^{-2}$ m

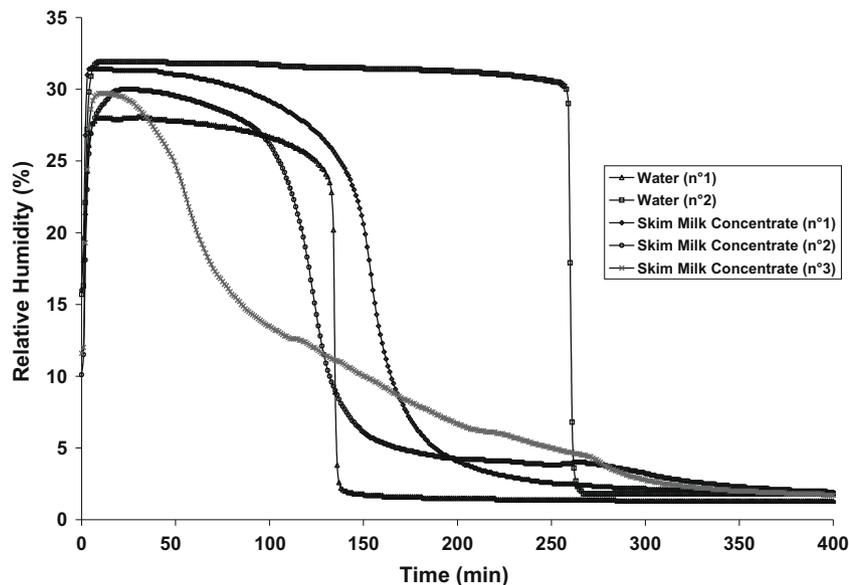**Fig. 1.** Equipment for drying by desorption.

Fig. 2. Drying by desorption. Relative humidity versus time.

Table 1
Total solids (theoretical, after oven drying or after desorption), water content and area under curve of water and skim milk concentrates

Sample	Theoretical total solids (%)	Total solids after oven drying (%)	Total solids after desorption (%)	Quantity of water before desorption (mg)	Area under curve (% min)	Extra energy/total energy (%)
Water (1)	–	–	–	80	3579	0.0
Water (2)	–	–	–	160	7688	0.0
Skim milk concentrate (No. 1)	30	28.5 ± 0.1	28.9 ± 0.7	112	5007	2.1
Skim milk concentrate (No. 2)	40	39.6 ± 0.1	40.8 ± 0.5	96	4355	8.6
Skim milk concentrate (No. 3)	50	49.7 ± 0.1	50.8 ± 0.4	80	3712	9.5

(about 160 mg) and the other containing an excess of a strong absorbent, in this case zeolite. The mass ratio is approx. 100 g zeolite to 1 g product. At 45 °C, the vapor pressure of zeolite is 30 Pa, whereas it is equal to the saturation vapor pressure (close to 3000 Pa) at the surface of the layer product. Water transfer takes place because of this vapor pressure gradient, in a similar way to the water transfer that occurs during drying. The main difference is in the kinetics of the transfer, which here takes 2 h compared to a few seconds during spray drying. This device was described by Schuck et al. (1998) (Fig. 1). One relative humidity (RH) sensor (Rotronic, Bassersdorf, Switzerland) over the two compartments makes it possible to plot RH in the cell as a function of time.

2.2. Chemical analysis

Total solids (TS) were determined on a 1.5 g sample mixed with sand after 7 h in a dry oven (105 °C).

2.3. Preparation of concentrate

Raw skim milk and sweet whey were provided by Triballat (Noyal sur Vilaine, France) and Euroserum (Montauban, France), respectively. Concentration of milk and whey was performed in a two-stage falling film vacuum evaporation pilot plant (GEA-Process Engineering, St. Quentin en Yvelines, France) at Bionov (Rennes, France) to obtain three skim milk concentrates (Nos. 1, 2 and 3 at 30%, 40% and 50% of TS, respectively) and a concentrated sweet whey at 50% of TS. The first evaporation stage was carried out at 60 °C and led to a concentrate temperature of 40 °C. The lactose of the concentrated sweet whey was crystallized up to 80%. The infant milk concentrate was reconstituted from infant formula milk powder (Laiterie de Montaigu, Montaigu, France) at 40% of TS. The glucose syrup was reconstituted from dried glucose syrup (dextrose equivalent 33) (Roquette Frères, Lestrem, France) at 40% of TS. Micellar casein isolate (MCI) at 19% of TS was separated from skim milk by tangential membrane microfiltration followed by purification through water diafiltration according to Fauquant et al. (1988), Pierre et al. (1992) and Schuck et al. (1994). Whey protein isolate (WPI) at 20% of TS was obtained by membrane tangential ultrafiltration and diafiltration (4 volumes) of microfiltrate, collected during MCI production with a spiral-wound organic membrane of 9.7 m² and a cutoff close to 20 kg mol⁻¹ at a volume reduction of 20. Finally, 38 dairy and food concentrates were obtained before drying by desorption (including variations in the concentrate temperature). All the analyses and experiments were performed in duplicate.

3. Results and discussion

3.1. Drying by desorption

The main aim of this study was to develop a method to simulate the transfer conditions (energy and water) of spray drying. Typical
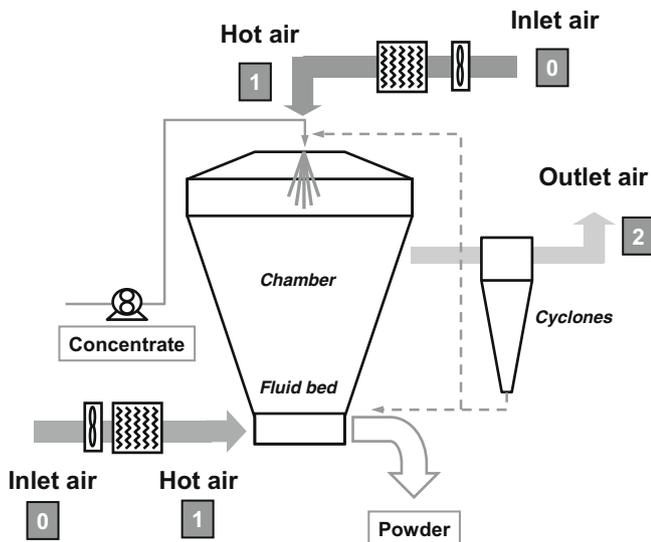

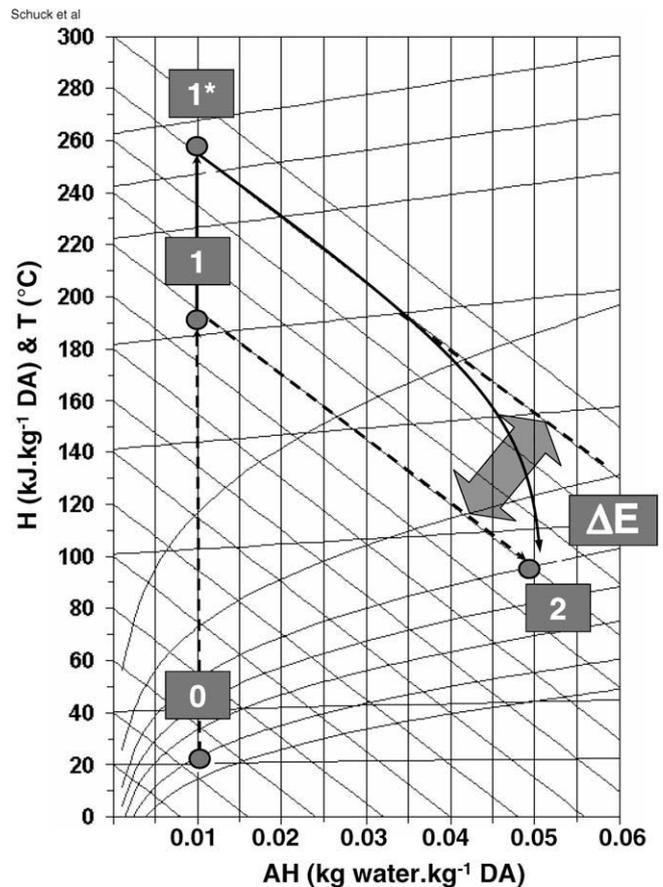**Fig. 3.** Diagram of a spray-drier including the sites of items 0, 1 and 2.

Fig. 4. Enthalpic Mollier diagram showing a theoretical (0, 1 and 2) and a real (0, 1* and 2) drying cycle.

curves (shown in Fig. 2) were registered with the water activity meter (Fig. 1), on pure water at two different sample weights (80 mg for water No. 1 and 160 mg for water No. 2) and on skim milk concentrates at different TS contents. These curves showed that the relative humidity (RH) from the pressure sensor as a function of time and as a function of skim milk concentrates and water can be represented by a sigmoid equation (Schuck et al., 1998):

$$RH = \frac{(\phi + \alpha t + \gamma \cdot t^2) + \delta}{\left[1 + \left(\frac{t}{\chi}\right)^\beta\right]^\varepsilon} \qquad (1)$$

where t is time of drying, ϕ, α and γ are the coefficients of the equation of the first part of the curves, δ is the final value for RH, χ is the inflection point, β is the slope of the sigmoidal part of the inflection point, ε is the asymmetry coefficient.

Two phases can be identified on the curves obtained for pure water and milk concentrates:

Initially, at the beginning of desorption, there was a constant RH (i.e. rate of drying) phase, which remained at approx. 30% whatever the product. This constant phase corresponds to free water evaporation. The second phase was the falling rate period, which was very short for pure water and much longer for milk concentrates, whatever the total solid content. We assumed that this corresponded to the evaporation of bound water, which involves extra energy (ΔE) in order to overcome binding (indirectly the bound strength of water). The area under the curve of each part thus represents the amounts of free and bound water desorbed, respectively. ΔE is calculated as a function of the drying kinetics according to the desorption curves (Fig. 2). When the drying kinetic of a concentrate is similar to the drying kinetic of pure water, no

Skim Milk Concentrate

	Mass flow rate (kg DA/h)	Enthalpy (kJ/kg DA)	Temperature (°C)	AH (g/kg DA)	RH (%)
Inlet air before heating		42.8	25	7	35.3
Inlet air after heating 'I'	80000	273.4	250	7	0.03
Cooling air 'C'	2500	42.8	25	7	35.3
Recirculation air 'R'	3000	42.8	25	7	35.3
Complementary air 'C'	1000	42.8	25	7	35.3
Air mix (I+C+R)	86500	256.1	231.8	7	0.04
Outlet air 1 stage (I+C+R+C)	86500	247.1	90.8	58.1	12
Integrated fluid bed inlet air 'B'	18000	108.4	88.9	7	1.7
Integrated fluid bed outlet air 'B'	18000	95.5	58.4	14	12
Overall outlet air (I+C+R+C+B)	104500	221	85.5	50.5	12.8

Evaporation capacity (kg/h)	4546.2	Wet bulb temperature of overall outlet air (°C)		49.1
Water flow rate in concentrate (kg/h)	4728	Dew temperature of overall outlet air (°C)		40.8
Concentrate flow rate (kg/h)	9649	Energy balance (kJ/kg water)		4151
Concentrate flow rate (l/h)	8040.8	Energy consumption ratio (60°C) (kg vapour/kg water)		1.8
Concentrate density (-)	1.2	Yield (60°C) (%)		56.8
Concentrate dry matter (%)	51	Cost ($/ton water)		69.2
Powder moisture (%)	4	Cost ($/ton powder)		63.8
Powder flow rate (kg/h)	5126	kWh cost ($)		0.06
Concentrate temperature (°C)	45			
Concentrate Cp (kJ/(kg.°C))	3.5			

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Fig. 5. Parameters of spray drying calculated by the SD2P® software.

ΔE is required. If the drying kinetic is two slower, energy requirement is two higher than for pure water. The difference with the latent heat of evaporation required for pure water determines ΔE.

The TS contents determined after oven drying and after drying by desorption are shown in Table 1. The values were very close compared to the theoretical findings although the standard deviations for drying by desorption were higher. The both before last columns of this Table show that the area under each curve for pure water and skim milk concentrates was related to the water content. The determination coefficient was close to 0.99. This correlation showed that drying by desorption can remove almost all the water whatever its nature (bound or free). The pure water sample (No. 1) and the skim milk concentrate (No. 3) had the same water content (80 mg) and similar areas under the curve (3579% min and 3712% min, respectively), but not the same drying time (about 150 min for pure water compared to 350 min for skim milk concentrate – see Fig. 2) and not the same extra energy (ΔE) compared to total energy (Table 1). The difference may be related to the change of diffusivity or availability of the water: the more bound the water, the longer the drying time. In first time, extra energy (ΔE) must be found to release the bound water physically and chemically, and then in second time, this released water could be evaporated by using the latent heat of evaporation, like pure water.

3.2. Drying parameters

To introduce our modeling approach, the known and unknown air parameters in a spray dryer are represented in Fig. 3. Three points are very important in characterization in terms of temperature (T, °C), relative humidity (RH, %) and absolute humidity (AH, kg water kg^{-1} dry air [DA]). Points 0, 1 and 2 correspond to the inlet air before heating, after heating (increase in T) and after drying (increase in AH), respectively. Air 0 characteristics can be easily determined by using a thermo-hygrometer. The AH of point 2 (AH_2) is given by dryer capacity, based on free water evaporation, and the RH of point 2 (RH_2) is given by the thermodynamic equilibrium between powder and outlet air (Schuck et al., 2005, 2008). This is mainly dependent on dryer configuration: for example, the RH_2 corresponding to $0.2a_w$ powder production is 10%, 15% and 20% in single, two and three-stage dryers, respectively. Outlet air temperature (T_2) is then obtained using the humid air chart. Finally, considering point 1, AH_1 is easily obtained from AH_0, consid-

ering either electric or gas (40 mg water kg^{-1} air °C^{-1}) heating. However, T_1 and RH_1 are unknown, except in the case of isenthalpic drying of free water (see Fig. 4), obtained in quite unrealistic conditions from an industrial point of view, by considering that spray drying is performed with no energy loss and by removing only free water. Comparing theoretical values and real values from dairy companies, Fig. 4 shows a significant difference between the theoretical (points 0, 1 and 2) and real values (points 0, 1* and 2). The difference between 1 and 1* in corresponding enthalpy should be directly correlated to extra energy ΔE, which is calculated as a function of the surface area under the curve of bound water desorbed according to the desorption curves (Fig. 2). Thus, the desorption method could take account of water availability.

The calculation method to determine ΔE was computerized to obtain new software ($SD^2P^{®}$, spray drying parameter simulation and determination) and registered at the APP (Association pour la Protection des Programmes) under the following identification IDDN.FR.001.480002.002.R.P.2005.000.30100. The next paragraph shows the relevance of this method.

Analysis of the desorption curve (Fig. 2) combined with knowledge of the temperature, total solids, density and specific heat capacity of the concentrate, air flow rates, theoretical water content in relation to water activity and RH_2, the current weather conditions (point 0), cost per kWh and the percentage of drying in the integrated fluid allowed determination of enthalpy, T, RH (of point 1 including ΔE) for each air, concentrate and powder flow rate, energy specific consumption, energy and mass balance, yield of the dryer and cost (in € or in $) to remove 1 kg of water or to produce 1 kg of powder. All these results are summarized in Fig. 5. This figure is a representation of the software delivery:

– in the upper part, air characteristics at the dryer/integrated fluid bed inlet and outlet, and
– in the lower part, flow, energy and cost calculations.

Fig. 6 represents the inlet air predicted as a function of the inlet air measured during 38 experiments (on skim milk, glucose syrup, WPI, MCI, infant formula milk and crystallized lactose whey) and on five types of spray dryer (from 5 kg h^{-1} to 6000 kg h^{-1}). The correlation was excellent, as were those calculated on the other parameters such as outlet air temperature, concentrate flow rate, powder flow rate and outlet air absolute humidity appearing in

$y = 0.99x$
$R^2 = 0.96$
$n = 38$

- Concentrated skim milk with (①②③) and without (❶❷❸) integration of ΔE
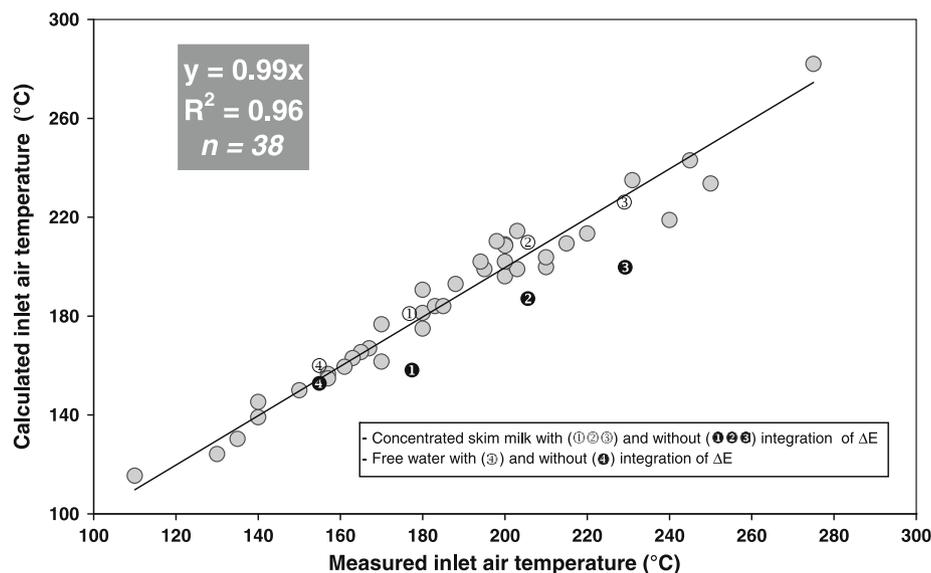- Free water with (③) and without (❹) integration of ΔE

Fig. 6. Correlation between calculated and measured inlet air temperature.

Table 2
Correlations between certain calculated and measured spray drying parameters

Drying parameters	R^2
Inlet air temperature (see Fig. 6)	0.96
Outlet air temperature	0.95
Concentrate flow rate	0.99
Powder flow rate	0.99
Outlet air absolute humidity	0.93

Table 2. The correlation is less good for the calculated inlet air temperature without integration of the ΔE except for pure water which has no required ΔE.

4. Conclusions

This study shows that drying by desorption is an excellent tool to determine the major spray drying parameters in relation to biochemical composition according to water availability, and to their desorption behavior (calculation of the ΔE). The experimental device differs from the spray drying installation in terms of duration of drying, temperature of drying, surface/volume ratio, etc. However, for these reasons some computational tools have been developed to improve the method by taking this into account. Validation tests (>30 products) indicated that this method could be applied to a wide range of food products and spray dryer types. For reasons of calculation speed and reliability, this method has been computerized and it can already be used in the determination of parameters of spray drying for food products. The name of the new software is "spray drying parameter simulation and determination software" ($SD^2P^®$) registered under the following identification: IDDN.FR.001. 480002.002.R.P.2005.000.30100.

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