Vol. 22, No. 2(2023) Alim237
Revista Mexicana de Ingeniería Química
Ingeniería de alimentos

Heat and mass transfer heterogeneous model applied for mathematical representation of Aloe vera extracts spray drying

Modelo heterogéneo de transferencia de calor y masa aplicado para la representación matemática del secado por aspersión de extractos de Aloe Vera

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Received: April 18, 2023; Accepted: June 15, 2023

Abstract
Punctual heterogeneous and homogeneous state-space dynamic models for mathematical representation of Aloe vera extracts spray drying were proposed. Punctual heterogeneous model considers 2 partial derivatives differential equations (PDDE) for particles in terms of Fourier's and Fick's laws, 2 ordinary differential equations (ODE) for drying air and 3 thermodynamic relations at interface. Homogeneous model considers 4 ODE for air and particles with the same 3 thermodynamic relations at interface. Punctual heterogeneous model was theoretically validated with divergence theorem. On the other way, fresh and concentrated Aloe vera extracts, obtained from plant expression (with 0.43% and 3.93% of total solids), were experimentally spray dried with 180-170 ºC, 100-80 ºC air input-output temperatures. Heterogeneous model solution produced a periodic chaotic dynamic behavior around homogeneous model results which moisture profile within 6.7-7.1 $\times 10^{-5}$ m diameter particles. Simulated outlet particle moistures reproduced moderately the experimental ones.
Keywords: spray drying, heat and mass transfer, homogeneous models, punctual heterogeneous models, Aloe vera extracts.

Resumen
Se proponen modelos dinámicos de espacio de estado heterogéneos puntuales y homogéneos para la representación matemática del secado por aspersión de extractos de Aloe Vera. El modelo heterogéneo puntual está formado por dos ecuaciones diferenciales en derivadas parciales (PDDE) para las partículas en términos de las leyes de Fourier y Fick, dos ecuaciones diferenciales ordinarias (ODE) para el aire de secado y tres relaciones termodinámicas en la interface. El modelo homogéneo está formado por 4 ODE para el aire de secado y partículas con las mismas tres relaciones termodinámicas en la interface. El modelo puntual heterogéneo fue validado teoréticamente con el teorema de la divergencia. Por otro lado, se secaron por aspersión extractos frescos y concentrados (0.43% y 3.93% de sólidos totales) de Aloe vera obtenidos por expresión de hojas de sábila a temperaturas de entrada y salida de aire de 170-180 ºC y 80-100 ºC respectivamente. El modelo puntual heterogéneo resultó en una conducta dinámica caótica alrededor del estado estable del modelo homogéneo, con perfil de humedad dentro de las partículas de 6.7-7.1 $\times 10^{-5}$ m de diámetro. Las humedades simuladas de salida de partícula reproducen las humedades experimentales con moderada aproximación.
Palabras clave: secado por aspersión, transferencia de calor y masa, modelos homogéneos, modelos heterogéneos puntuales, extractos de Aloe vera.

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https://doi.org/10.24275/rmiq/Alim237
ISSN:1665-2738, *issn-e*: 2395-8472

1 Introduction

Spray drying has been one of the most important unit operations in food engineering. It has been applied in commercial scale since the firs middle of century XX (Seltzer and Settelmayer, 1949). Practically, all powder foods in bulk are produced by spray drying (Straatsma *et al*., 1999; Razmi *et al*., 2021) and, it is important to remark, it is a high energy demand operation. Spray drying processes performed at 25% of thermal efficiency, require 4 times of water latent heat (around 4 x 2176 kJ/kg evaporated water at 70 °C) plus mechanical work for product and air pumping (Aguirre *et al*., 2019; Rodríguez-Jimenes *et al*., 2014; Villegas-Santiago *et al*., 2020). Therefore, the application of mathematical modeling with rigorous thermodynamic (reversible and irreversible) approaches is fundamental for to increase its thermal efficiency (Aguirre-Alonso *et al*., 2019; Villegas-Santiago *et al*., 2020). Rigorous momentum transfer modeling by applying CFD is also fundamental for the geometric design of drying chamber (Frydman *et al*., 1999; Straatsma *et al*., 1999; Langrish, 2009; Jubaer *et al*., 2018; Razmi *et al*., 2021). However, the design of drying chamber volume and drying air requirements (and therefore energy consumption) may be performed with averaged heat and mass balances assumed drying chamber as an ideal mixed unit (Palencia *et al*., 2002; Luna-Solano *et al*., 2005; Aguirre-Alonso *et al*., 2020). Independently of the mathematical model applied (CFD, ideal mixed units or both), the particle-gas interface may be represented with homogeneous or heterogeneous models. Homogeneous model represents heat and mass transfer at interface with convection equations in gas phase side, assuming that particle temperature and moisture is equal to interface values; which implies that the complete mathematical model is represented in a homogeneous gas phase (Aguirre-Alonso *et al*., 2020). Heterogeneous model represents the heat and mass transfer at interface as a continuous phenomenon between convection in gas phase side and conduction-diffusion in particle side. Heterogeneous models can be lumped, when conduction-diffusion are represented with a volume integrated model (Palencia *et al*., 2002; Luna-Solano *et al*., 2005); or punctual, when conduction-diffusion are represented with Fourier's and Fick's laws within particles and considering that there is a continuous replacement of energy and moisture in particles. Palencia *et al*., (2002) showed that lumped heat and mass transfer in particles joint with heat and mass balances in air drying, assuming complete mixing within drying chamber, resulted in a state-space dynamic model that reproduce experimental spray drying dynamics. Luna-Solano *et al*., (2005) solved the state-space dynamic model, proposed by Palencia *et al*., (2002), until steady state for yeast spray drying optimization. Even the models which consider momentum, heat and mass transfer (Frydman *et al*., 1999; Straatsma *et al*., 1999; Langrish, 2009; Judbaer *et al*., 2018; Razmi *et al*., 2021) are homogeneous or lumped heterogeneous models. Therefore, in our knowledge, spray drying mathematical models, that considers Fourier's and Fick's laws within particles with a continuous replacement of energy and moisture have not been reported.

On the other hand, Aloe vera has been raised in attention due to its applications in different fields. As example, Aloe vera has been used for several medical applications. As example, wound healing, anti-inflammatory, anticancer, antidiabetic, antihyperlipidemic, antioxidant, dental protector, asthma relieve, laxative effect, treatments against STD diseases and other more (Reynolds & Dweck, 1999; Talmadge *et al*., 2004; Chow *et al*., 2005; Alvarado-González *et al*., 2012; Domínguez-Fernández *et al*., 2012; Aslam-Maan *et al*., 2018). In the food industry Aloe vera has had applications as nutritional and functional food, antimicrobial agent and as an edible coating (McAnalley 1993; Hamman 2008; Domínguez-Fernández *et al*., 2012; Cervantes-Martínez *et al*., 2014). Aloe vera solids are composed mainly by 55 % of polysaccharides, 17 % of sugars and 1 % of phenolic compounds (Chow *et al*., 2005; Hamman 2008; Domínguez-Fernández *et al*., 2012). The polysaccharide with the highest concentration is acemannan, which represents 60% of the polysaccharides present in aloe (Chow,*et al*., 2005; Talmadge *et al*., 2004). Due to its high content of polysaccharides, Aloe vera has also been used as a support material or encapsulating material in spray drying (Medina-Torres *et al*., 2016; Medina-Torres *et al*., 2019a; Medina-Torres *et al*., 2019b; Ceja-Media *et al*., 2021). Aloe vera extracts obtained by expression has high water content (99-98 %) and therefore evaporation and spray drying unit operations are required for aloe vera powder production. Exist some research on Aloe vera spray drying (Cervantes-Martínez *et al*., 2014; Medina-Torres *et al*., 2016; Medina-Torres *et al*., 2019a; Medina-Torres *et al*., 2019b; Ceja-Media *et al*., 2021; Sharma *et al*., 2022) but the whole of referred studied are focused in rheological properties of reconstituted powder, in their functional properties or in applications as encapsulating material. In our knowledge the mathematical modeling of Aloe vera extracts spray drying with rigorous thermodynamic models has not been reported.

Therefore, the scientific hypothesis planted in the present research is that a punctual heterogeneous model in terms of Fourier's and Fick's law within particles with continuous replacement of energy and moisture, represent rigorously the heat and mass transfer phenomena during spray drying process. Such hypothesis was theoretically validated, with application of divergence theorem, and experimentally validated with fresh and concentrated Aloe vera extracts spray drying at different conditions. Both models (punctual heterogeneous and homogeneous) were applied for simulation of high energy efficiency Aloe vera extracts spray drying.

2 Modeling

2.1 *Spray drying homogeneous modeling*

Spray drying is a simultaneous heat and mass transfer process in which heat is transferred from a gas phase to a spread liquid phase which is transformed in solid phase when is dried. This liquid phase transformed in solid phase will be called particles phase. For any processes, state-space is the set of differential equations that define the state variables over a given time dominion (Palencia *et al*. 2002). It has been theoretically and experimentally demonstrated (Palencia *et al*., 2002; Luna-Solano *et al*., 2005; Aguirre-Alonso *et al*., 2019) that spray driers may be represented in the state-space assuming ideal mixing in the drying chamber, taking air and particles temperatures joint with air and particles moisture as state variables. Ideal mixing assumption implies that the referred four state variables are uniformly distributed in drying chamber and convection terms may considered in global air and particles flows. Other researches (Straatsma *et al*., 1999; Langrish, 2009; Jubaer *et al*., 2018; Razmi *et al*., 2021) which proposed mathematical modeling with different mix patterns and momentum equations with computational fluids dynamics (CFD) did not show experimental evidence that the finer scale modeling produces a better approximation to experimental results. Additional problem with CFD is that requires special software not available charge-free but it is important to remark, that CFD is fundamental for drying chamber geometrical design (Razmi *et al*., 2021).

Therefore, assuming ideal mixed drying chamber, the application of non-steady thermodynamic first law based enthalpy balance for the open heterogeneous system (Balzhiser *et al*., 1972) of air (γ-phase)-particles (β-phase), in the spray dryer chamber with continuous air and particles flows (G_γ and G_β), the following equations are obtained,

$$\rho_\gamma V\varepsilon\frac{dh_\gamma}{dt} = -G_\gamma(h_\gamma - h_{\gamma 0}) + (q_{\beta\gamma} + q_{wv})a_{\beta\gamma}V - q_{env}, \quad for\ t > 0 \quad (1)$$

$$\rho_\beta V(1-\varepsilon)\frac{dh_\beta}{dt} = -G_\beta(h_\beta - h_{\beta 0}) - (q_{\beta\gamma} + q_{wv})a_{\beta\gamma}V, \quad for\ t > 0 \quad (2)$$

Where input ($h_{\gamma 0}, h_{\beta 0}$), output ($h_\gamma, h_\beta$), gas and particles enthalpies respectively, are,

$$h_\gamma = Cp_aT_\gamma + (h^0_{wv} + Cp_{wv}T_\gamma)X_\gamma \quad (3)$$

$$h_\beta = (Cp_s + Cp_wX_{w\beta})T_\beta \quad (4)$$

$q_{\beta\gamma}$ is the heat flux transferred by convection from air phase to particles phase due to temperature gradient,

$$q_{\beta\gamma} = U_\gamma(T_i - T_\gamma) \quad (5)$$

q_{wv} is the heat required for water evaporation,

$$q_{wv} = N_{\beta\gamma}\lambda_w \quad (6)$$

$N_{\beta\gamma}$ is the mass transfer flux (evaporated water) by convection at the particle-gas interface,

$$N_{\beta\gamma} = k_{c\gamma}\rho_\gamma(X_{\gamma i} - X_\gamma) \quad (7)$$

and q_{env} is the heat lost by convection to environmental surrounding dryer chamber,

$$q_{env} = U_{env}A_{env}(T_\gamma - T_{env}) \quad (8)$$

q_{env} and outlet total enthalpy ($G_\gamma h_\gamma$ plus $G_\beta h_\beta$) are dissipated to environmental and therefore, transformed in surroundings entropy in agree with thermodynamic second law (Balzhiser *et al*., 1972). In Eqs. (1) and (2) the pressure change ($\Delta p/\rho$) term is considered as neglected with respect to heat transfer ($q_{\beta\gamma}$, q_{wv}) terms. The four state variables: air temperature (T_γ), particle temperature (T_β), air moisture (X_γ) and particle moisture (X_β), can be identified in Eqs. (1)-(8). The rest of variables are defined in nomenclature section. Eqs. (1) and (2) are two differential equations of state-space, the other two are obtained from non-steady water mass balance for the same heterogeneous system in the ideally mixed spray dryer chamber,

$$\rho_\gamma V\varepsilon\frac{dX_\gamma}{dt} = -G_\gamma(X_\gamma - X_{\gamma 0}) + N_{\beta\gamma}a_{\beta\gamma}V, \quad for\ t > 0 \quad (9)$$

$$\rho_\beta V(1-\varepsilon)\frac{dX_\beta}{dt} = -G_\beta(X_\beta - X_{\beta 0}) - N_{\beta\gamma}a_{\beta\gamma}V, \quad for\ t > 0 \quad (10)$$

Particles-gas interface must be in thermal and mass equilibrium. Thermal equilibrium implies that both interface sides are at the same temperature (T_i). Water mass equilibrium implies that moisture in gas phase side ($X_{\gamma i}$) must be in equilibrium with the correspondent moisture by particle side ($X_{\beta i}$). As both moistures are expressed in mass relation (kg water/kg dry matter), $X_{\gamma i}$ is calculated from Raoult's law corrected by water thermodynamic activity (a_w) in order to considerate non-ideal product-water mixtures (Palencia *et al*., 2002; Langrish, 2009),

$$X_{\gamma i} = \frac{a_wp^0_w/p}{1 - a_wp^0_w/p}\frac{M_w}{M_a} \quad (11)$$

where a_w as function of product moisture ($X_{\beta i}$) at a given T_i is known as sorption isotherm. A general model for sorption isotherm is (Palencia *et al*., 2002),

$$a_{wi} = 1 - \exp(-k_1T_i^{k_2}X_{\beta i}^{k_3}) \quad (12)$$

The water vapor pressure (p^0_w) can be represented by extended Antoine's equation (Perry *et al*., 1999),

$$p^0_w = \exp\left(73.649 - \frac{7258.2}{T_i} - 7.3037\ln T_i + 4.1653\times 10^{-6}T_i^2\right) \quad (13)$$

In the particular case of very small particles (in the magnitude order of 1.0× 10^{-5} m or smaller) it may be assumed that particles bulk temperature and moisture can be considered equal to surface ones ($T_\beta = T_i$ and $X_\beta = X_{\beta i}$). Therefore, in such particular case, Eqs. (1)-(13) are completely defined in terms of gas phase and particle surface and Eqs. (1)-(2), (9)-(10) complemented with (3)-(8) and (11)-(13) are the state-space equations for the homogeneous model.

2.2 *Spray drying punctual heterogeneous modeling*

Homogeneous model is the state-space assuming very small particles. A general model must considerate the effect of heat conduction and moisture diffusion within particles phase. Palencia *et al*., (2002) and Luna-Solano *et al*., (2005) considered internal conduction-diffusion in a lumped model, that is with pseudo internal heat and mass transfer coefficients in terms of heat conduction and mass diffusion. In present paper, a rigorous and not previously proposed (at least in our knowledge) heat conduction and water diffusion model, is proposed.

Heat conduction and moisture diffusion driving forces are heat ($q_{\beta\gamma}$) and mass ($N_{\beta\gamma}$) transfer fluxes at interface. Assuming spherical particles uniformly distributed in drying chamber with an average radius (R), the difference between the heat transferred from air to particles (Eq. 5) and the heat applied in water evaporation (Eq. 6) at air side interface produces the temperature gradient at particle side interface resulting in heat conduction expressed in 1D spherical coordinates as,

$$-k_\beta \frac{\partial T_\beta}{\partial r} = q_{\beta\gamma} + q_{wv}, \quad \text{at } r = R \text{ and } t > 0 \qquad (14)$$

For mass transfer the mass (evaporated water) transferred from particles to air (Eq. 7) at air side interface produces the moisture gradient at particle side interface resulting in water diffusion expressed in 1D spherical coordinates as,

$$-D_{w\beta}\rho_\beta \frac{\partial X_\beta}{\partial r} = N_{\beta\gamma}, \text{ at } r = R \text{ and } t > 0 \qquad (15)$$

Eqs. (14)-(15) represent the boundary conditions for Fourier's and Fick's laws within particles. However, heat conduction and moisture diffusion within particles must consider that the process is continuous. That is, exists a continuous inlet-outlet of particles to drying chamber and therefore the particles are continuously replaced with new particles with contain energy and moisture. Therefore, Fourier's and Fick's laws within particles must be written in 1D spherical coordinates as,

$$\rho_\beta Cp_\beta \frac{\partial T_\beta}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k_\beta \frac{\partial T_\beta}{\partial r}\right) - \frac{G_\beta Cp_\beta}{(1-\varepsilon)V}(T_\beta - T_{\beta 0})$$
$$\text{for } 0 < r < R \text{ and } t > 0 \quad (16)$$

$$\frac{\partial X_\beta}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D_{w\beta} \frac{\partial X_\beta}{\partial r}\right) - \frac{G_\beta}{\rho_\beta(1-\varepsilon)V}(X_\beta - X_{\beta 0})$$
$$\text{for } 0 < r < R \text{ and } t > 0 \quad (17)$$

Eqs. (16) and (17) take into account, in non-steady state, the average conduction and diffusion within particles (in 1D radial spherical coordinate) and the continuous replacement of energy (considered and averaged specific heat Cp_β) and moisture. Eqs. (16) and (17) may reach a steady state when the temperature and moisture internal profiles in particles produces a heat conduction and moisture diffusion equal to inlet-outlet energy and moisture. Therefore, the proposed punctual heterogeneous state-space for spray drying process is formed by Eqs. (1), (9), (16) and (17) with Eqs. (14) and (15) as interface boundary conditions and complemented with Eqs. (3)-(8), (11)-(13).

3 Methodology

3.1 Model solution

3.1.1 Homogeneous model

Eqs. (1)-(13) were solved until steady-state by using ode15s MatLab routine. MatLab is a popular and powerful low-cost software, available at special cost for student in which ode15s is a routine of basic module. Eqs. (3)-(4) was substituted in Eqs. (1) and (2) in order to obtain ODEs explicit in air and particle temperatures (T_γ and T_β). Initial conditions were taken equal to air and product moisture and temperature inlet values, that is $T_\gamma(t = 0) = T_{\gamma 0}$, $T_\beta(t = 0) = T_{\beta 0}$, $X_\gamma(t = 0) = X_{\gamma 0}$ and $X_\beta(t = 0) = X_{\beta 0}$. Interface variables were taken equal to particle variables, that is, $T_i = T_\beta$ and $X_i = X_\beta$ in agree with fundamental assumption for homogeneous model (particle radius is the magnitude order of 1.0×10^{-5} m or smaller).

The particle radius was estimated from Nukiyama-Tanasawa Equation for spray nozzles (Perry *et al*., 1997),

$$2R = \frac{1920\sqrt{\alpha}}{v_{\beta\gamma}\sqrt{\rho_{\beta l}}} + 597\left(\frac{\mu_{\beta l}}{\sqrt{\alpha\rho_{\beta l}}}\right)^{0.45}\left(\frac{1000 Q_{\beta l}}{Q_\gamma}\right)^{0.45} \qquad (18)$$

Assuming spherical particles with average radius calculated from Eq. (18), the particles specific surface is,

$$a_{\beta\gamma} = \frac{3(1-\varepsilon)}{R} \qquad (19)$$

Heat and mass transfer coefficients (U_γ and $k_{c\gamma}$) around particles were estimated from Ranz and Marshal Eqs. joint with Colburn correlation (Bird, 1960) around spheres,

$$Nu = 2.0 + 0.6Re^{0.5}Pr^{0.33} \qquad (20)$$

$$Sh = 2.0 + 0.6Re^{0.5}Sc^{0.33} \qquad (21)$$

Where $Nu = U_\gamma\phi_\beta/k_\gamma$, $Sh = k_{c\gamma}\phi_\beta/D_{w\gamma}$, $Pr = Cp_\gamma\mu_\gamma/k_\gamma$, $Sc = M_\gamma/(\rho_\gamma D_{w\gamma})$. Eqs. (20)-(21) were previously applied in order to estimate heat and mass transfer coefficients around particles during spray drying by Straatsma *et al*., (1999), Palencia *et al*., (2002), Langrish, (2009), Jubaer *et al*., (2018) and Aguirre-Alonso *et al*., (2019).

Air density (ρ_γ) were calculated with ideal gases law, public domain data for air viscosity (μ_γ) and heat conductivity (k_γ) were fitted to following equations,

$$\mu_\gamma = 4.249136\times 10^{-8}(T_{\gamma K} + 273.15) + 5.8652\times 10^{-6} \qquad (22)$$

$$k_\gamma = 8.40441\times 10^{-5}(T_{\gamma K} + 273.15) + 4.629375\times 10^{-5} \qquad (23)$$

Water vapor diffusivity in air was estimated from Chapman-Enskog Eq. (Poling *et al*., 2001),

$$D_{w\gamma} = \frac{2.6\times 10^{-7}\sqrt{(T+273.15)^3\left(\frac{1}{M_w}+\frac{1}{M_a}\right)}}{2p\sigma_{w\gamma}^2\Omega_D} \qquad (24)$$

for p in , $\sigma_{w\gamma} = (\sigma_w + \sigma_\gamma)/2$, $\varepsilon_{w\gamma} = \sqrt{\varepsilon_w\varepsilon_\gamma}$ and

$$\Omega_D = \frac{1.06036}{T^{*0.1561}} + \frac{0.193}{\exp(0.47635T^*)} + \frac{1.03587}{\exp(1.52996T^*)} + \frac{1.76474}{\exp(3.89411T^*)} \qquad (25)$$

Where $T^* = k(T + 273.15)/\varepsilon_{w\gamma}$.

With $\sigma_w = 2.641$, $\sigma_\gamma = 3.711$, $\varepsilon_\gamma/k = 78.6$, $\varepsilon_w/k = 809.1$ for water and air (Poling *et al*., 2001).

Table 1. Experimental design for fresh and concentrated Aloe vera extract spray drying.

Run	$T_{\gamma 0}$ (ºC)	$T_{\gamma 1}$ (ºC)	$x_{\beta s0}$	$X_{\gamma 0}$ (gg^{-1})
1	180	90	0.0043	0.017
2	180	80	0.0043	0.017
3	170	90	0.0043	0.016
4	170	90	0.0043	0.017
5	180	80	0.0043	0.016
6	180	90	0.0043	0.016
7	170	98	0.0393	0.02
8	170	103	0.0393	0.019

3.1.2 Pointwise heterogeneous model

Eqs. (1)-(8), and (11)-(17) were solved until steady-state joint with Eqs. (18)-(25) for particle radius estimation and transfer coefficients. In order to obtain a state-space, Eqs. (16)-(17) were space discretized by first order finite differences,

$$\frac{dT_{\beta j}}{dt} = \frac{k_\beta\left(\frac{r_{j+1/2}^2(T_{\beta j+1}-T_{\beta j})}{\Delta r} - \frac{r_{j-1/2}^2(T_{\beta j}-T_{\beta j-1})}{\Delta r}\right)}{\rho_\beta C p_{\beta j} r_j^2 \Delta r} - \frac{G_\beta}{\rho_\beta(1-\varepsilon)V}(T_{\beta j} - T_{\beta 0}) \tag{26}$$

$$\frac{dX_{\beta j}}{dt} = \frac{D_{w\beta}\left(\frac{r_{j+1/2}^2(X_{\beta j+1}-X_{\beta j})}{\Delta r} - \frac{r_{j-1/2}^2(X_{\beta j}-X_{\beta j-1})}{\Delta r}\right)}{r_j^2 \Delta r} - \frac{G_\beta}{\rho_\beta(1-\varepsilon)V}(X_{\beta j} - X_{\beta 0}) \tag{27}$$

for $r_j = j\Delta r$; $\Delta r = R/N$ and $j = 1,2,\ldots,N$.

At $j = N$ Eqs. (26)-(27) must be solved simultaneously with discretized interface boundary conditions (Eqs. 14-15),

$$T_{\beta N+1} - T_{\beta N-1} = -\frac{q_{\beta\gamma} + q_{wv}}{k_{\beta N}} 2\Delta r \tag{28}$$

$$X_{\beta N+1} - X_{\beta N-1} = -\frac{N_{\beta\gamma}}{\rho_{\beta N} D_{w\beta N}} 2\Delta r \tag{29}$$

Eqs. (1)-(8), (11)-(15) and (26)-(29) is a set of $2N + 2$ ODEs which represent the proposed state-space punctual heterogeneous model applied for rigorous mathematical modeling of spray drying process supported in reversible (enthalpy balance and equilibrium relations) and irreversible (heat and mass transfer conduction-diffusion, heat losses to environmental and not-recovery output thermal energy) thermodynamics principles. The referred ODEs were solved until steady state with ode15s MatLab routine taking inlet variables as initial conditions, that is $T_\gamma(t = 0) = T_{\gamma 0}$, $T_{\beta j}(t = 0) = T_{\beta 0}$ for $j = 1,2,\ldots,N$, $X_\gamma(t = 0) = X_{\gamma 0}$ and $X_{\beta j}(t = 0) = X_{\beta 0}$ for $j = 1,2,\ldots,N$. In order to validate the proposed model, experimental assays of Aloe vera leaves extracts spray drying were performed.

3.2 Experimental

Aloe vera leaves, used as raw material, were obtained from local market in Paso de Ovejas, Veracruz, México. Aloe vera gel was obtained by cutting the leaves and squeezing. Resulting gel was frozen at -5 °C and thawed at 28 °C until experiments. Aloe vera extracts were de-frozen at environmental conditions and filtered through Whatman No. 1 paper which retains particles larger than 11 μm. Aloe vera extracts moisture content were evaluated in vacuum oven resulting in 99.57 % on wet basis. Some fresh Aloe vera extracts (99.57% moisture) were vacuum evaporated until 3.8% of solids. Fresh and evaporated extracts were spray dried in a laboratory spray dryer Büchi B-191 at 180-170 ºC for drying air inlet temperature by adjusting feed flow in order to obtain 80-100 ºC in drying air outlet temperature. In order to estimate drying air flow the air velocity in air outlet duct (R_1 = 0.01875 radius) was measurement with a digital anemometer. Each spray drying treatment was performed by duplicate. The complete experimental runs were listed in Table 1, which are numbered in execution order. Runs 1-6, 2-5, 3-4 and 7-8 are replicates and three samples of dried product were taken for outlet moisture evaluation in vacuum oven.

4 Results and discussions

4.1 Theoretical validation

Lumped heterogeneous models have been previously applied (Palencia *et al.*, 2002; Luna-Solano *et al.*, 2005). The proposed punctual heterogeneous model can be validated, if it can be reduced to a previous lumped heterogeneous model. Lumped models represent the process in terms of particle bulk variables $\langle T_\beta\rangle = \int\int\int_{V_\beta} T_\beta\ dV/V_\beta$ and $\langle X_\beta\rangle = \int\int\int_{V_\beta} X_\beta\, dV/V_\beta$. Integrating Eqs. (16) and (17) over particles volume and applying the divergence theorem,

$$\rho_\beta C p_\beta \frac{\partial\langle T_\beta\rangle}{\partial t} = \int\int_{A_{\beta\gamma}} k_\beta \frac{\partial T_i}{\partial r} dA_{\beta\gamma} - \frac{G_\beta C p_\beta}{(1-\varepsilon)V}(\langle T_\beta\rangle - \langle T_{\beta 0}\rangle) \tag{30}$$

$$\frac{\partial\langle X_\beta\rangle}{\partial t} = \int\int_{A_{\beta\gamma}} D_{w\beta} \frac{\partial X_{\beta i}}{\partial r} dA_{\beta\gamma} - \frac{G_\beta}{\rho_\beta(1-\varepsilon)V}(\langle X_\beta\rangle - \langle X_{\beta 0}\rangle) \tag{31}$$

The integral of Eqs. (30)-(31) right side represent the entire heat and mass transfer through particle-air interface which in agree to Eqs. (14)-(15) are,

$$-\int\int_{A_{\beta\gamma}} k_\beta \frac{\partial T_i}{\partial r} dA_{\beta\gamma} = (q_{\beta\gamma} + q_{wv}) a_{\beta\gamma} V \quad (32)$$

$$-\int\int_{A_{\beta\gamma}} D_{w\beta}\rho_\beta \frac{\partial X_{\beta i}}{\partial r} dA_{\beta\gamma} = N_{\beta\gamma} a_{\beta\gamma} V \quad (33)$$

Additionally, integrals (32)-(33) may be represented as a lumped heat and mass transfer in terms of apparent heat and mass transfer coefficients in particles (U_β and $k_{c\beta}$),

$$-\int\int_{A_{\beta\gamma}} k_\beta \frac{\partial T_i}{\partial r} dA_{\beta\gamma} = -U_\beta \left(T_i - \langle T_{\beta 1}\rangle\right) a_{\beta\gamma} V \quad (34)$$

$$-\int\int_{A_{\beta\gamma}} D_{w\beta}\rho_\beta \frac{\partial X_{\beta i}}{\partial r} dA_{\beta\gamma} = -k_{c\beta}\rho_\beta \left(X_{\beta ii} - \langle X_{\beta 1}\rangle\right) a_{\beta\gamma} V \quad (35)$$

Where the pseudo heat and mass transfer coefficients are defined, from the analytical solution of general heat and mass transfer equations in a media in contact with a finite volume fluid phase (Vargas-González, *et al*., 2017), as,

$$U_\beta = \frac{\Omega k_\beta}{R\theta} \quad (36)$$

$$k_{c\beta} = \frac{\Omega D_{w\beta}}{R\theta} \quad (37)$$

With topological and geometric factors for spheres are $\Omega = \pi^2$ and $\theta = 3$. Eqs. (30)-(31) with (34)-(35) imply,

$$U_\beta \left(T_i - \langle T_{\beta 1}\rangle\right) = -U_\gamma (T_i - T_\gamma) - k_{c\gamma}\rho_\gamma (X_{\gamma i} - X_{\gamma 1})\lambda \quad (38)$$

$$k_{c\beta}\rho_\beta (X_{\beta i} - \langle X_{\beta 1}\rangle) = -k_{c\gamma}\rho_\gamma (X_{\gamma i} - X_\gamma) \quad (39)$$

Eqs. (1)-(8), (11)-(13), (30)-(31) with (34)-(35) and (38)-(39) represent the heterogeneous lumped models that was previously reported by (Palencia *et al*., 2002 and Luna-Solano *et al*., 2005) which theoretically validates the main research hypothesis: Eqs. (1)-(8) and (11)-(17) represents a valid punctual heterogeneous model which was not previously published.

Punctual heterogeneous models represent the heat and mass transfer phenomena in an averaged media contacting a fluid phase. Lumped models approximate the punctual ones, in some cases with poor performance (Vargas-González *et al*., 2017). The lumped model advantage is that they are 4 ODE in simultaneous with three algebraic equations (11, 38-39); instead the punctual model are two ODE (Eqs.1-2) and two PDDE (Eqs. 16-17 with 14-15 as boundary conditions). However, the three algebraic equations of lumped model form a non-linear system due the complex nature of Eq. (11) which must be solved simultaneously with Eqs. (38)-(39) in order to evaluate $X_{\beta i}$, $X_{\gamma i}$ and T_i. This implies that numerical solution of the ODE system (1)-(2), (30)-(31) must be solved applying a non-linear solution algorithm for the three algebraic Eqs. (11), (38)-(39) in each ODE solution step. Palencia *et al*., (2002) and Luna-Solano *et al*., (2005) reported the solution complexity, joint with chaotic behavior of solution. Otherwise, PDDE punctual model may be solved by space discretizing Eqs. (16)-(17) in N nodes (Eqs. 26-29) within particles for to obtain a $2N + 2$ ODE system, which can be solved with ordinary algorithms for ODE avoiding the algebraic non-linear system. Both mathematical modeling approaches, homogeneous model (Eqs. 1-15 assuming $T_\beta = T_i$, $X_\beta = X_{\beta i}$) and punctual heterogeneous model were solved until steady-state and compared with experimental results of Aloe vera extract spray drying.

4.2 Experimental validation

Experimental spray drying was operated keeping constant the air velocity in $v_{\gamma 1}$ = 13.4 m s^{-1} at air output duct with R_1 = 0.01875 radius. Therefore, air flow was calculated with,

$$G_\gamma = \pi R_1^2 v_{\gamma 1}\rho_{\gamma 1} \quad (40)$$

Where air density ($\rho_{\gamma 1}$) was estimated with ideal gas law at averaged air outlet-inlet temperatures. The general variables and properties applied in models were listed in Table 2. Fresh and concentrated Aloe vera extract feed flows ($Q_{\beta 1}$) were experimentally adjusted in order to reach the outlet desired temperature (Table 1) and the results are listed in Table 3. Inlet feed flow (G_β) and water mass relation ($X_{\beta 0}$) used in models were calculated from Tables 1 and 3 values as is indicated at follow,

$$X_{\beta 0} = \frac{1 - x_{\beta s}}{x_{\beta s}} \quad (41)$$

$$G_\beta = \frac{Q_{\beta l}}{3600}\frac{\rho_{\beta 0}}{1000} x_{\beta s} \quad (42)$$

Table 2. General properties applied in Eqs. (1)-(17).

Property or variable	Value	Units (ºC)
$Cp_a, Cp_s, Cp_{wv}, Cp_w$	1000, 1650, 1800, 4185	J kg^{-1} K^{-1}
k_1, k_2, k_3	3.86, 0.0, 0.985^a	Eq. (12) T in K
h_{wv}^0	2.5×10^6	J kg^{-1}
ε	0.995	
λ	$h_{vw}^0 + Cp_{vw}T - Cp_wT$	J kg^{-1}
$D_{w\beta}$	10^{-10}	m^2 s^{-1}
k_β	0.2^b	W m^{-1} K^{-1}
$\rho_{\beta 0}$	1059.4^a	kg m^{-3}
V	4.2×10^{-3}	m^3
A_{env}	0.19^c	m^2

a: Fitted Eq. (12) to Vega et al., (2007) data.
b: Aguirre-Alonso et al., (2019).
c: Experimental device.

Table 3. Experimental and simulated results for concentrated Aloe vera extract spray drying

Run[a]	$Q_{\beta l}$[b] (L h^{-1})	$X_{\beta 1 exp}$[c] (g g^{-1})	$X_{\beta 1 sim}$[d] (g g^{-1})	η%	U_{env}[e] (W m^{-2} K^{-1})
180-90	0.3597	0.065±0.006	0.018	12.4	77
180-80	0.467	0.13±0.0005	0.029	16.3	97
170-90	0.2421	0.015±0.001	0.013	8.9	74
170-90	0.2596	0.027±0.003	0.016	9.5	74
180-80	0.378	0.14±0.01	0.026	13.2	105
180-90	0.3392	0.055±0.002	0.017	11.8	79
170-98	0.106	0.045±0.004	0.011	3.68	65
170-103	0.108	0.045±0.0009	0.0085	3.74	57

a: $T_{\gamma 0}$-$T_{\gamma 1}$ with respect to Table 1.
b: Adjusted in order to obtain the output temperature ($T_{\gamma 1}$) in the experimental device.
c: Mean of three replicates ± 95% confidence interval variation.
d: Averaged over particle volume of the averaged last 10 times.
e: Adjusted in order to obtain experimental $T_{\gamma 1}$ in simulation.

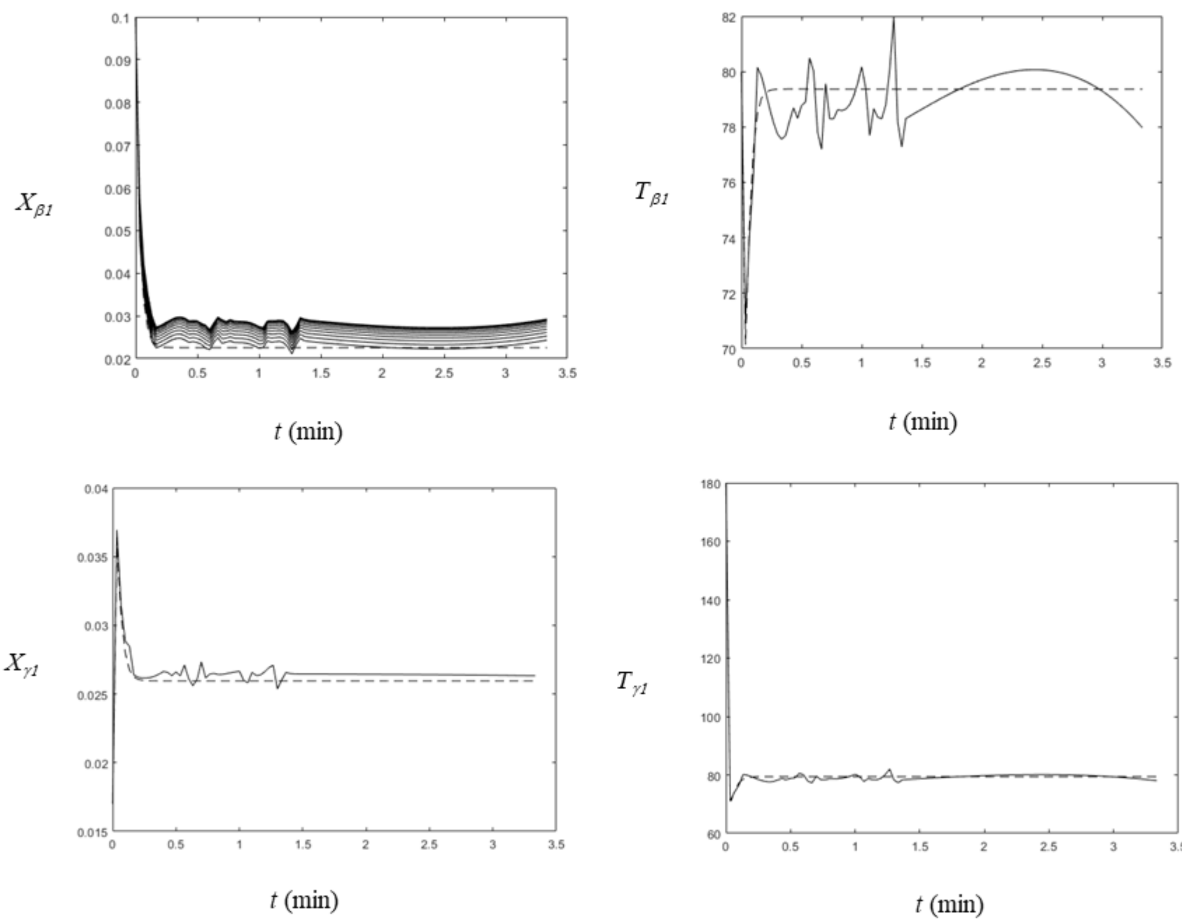

Fig. 1. Aloe vera spray drying dynamic simulation with punctual heterogeneous (—) and homogeneous (- - -) model at run 5 conditions of Tables 1-3.

Due to environmental variable conditions in laboratory and the stochastic nature of heat transfer coefficient from dryer chamber to environmental (U_{env}) observed in preliminary experimental test (with water and with Aloe vera extracts as feed flow), U_{env} was adjusted in simulations in order to obtain the experimental outlet temperature. Water molecular diffusion of the entire Table 1 treatments were between (2-3)× 10^{-5} m^{2}s^{-1} (Eqs. 24-25); mass transfer coefficients between 1.5-1.7 ms^{-1} (Eqs. 21-22); heat transfer coefficients between (1.4-1.8)× 10^3 W m^{-2}K^{-1} (Eqs. 20, 22-23); droplet diameter between (6.5-7.0)× 10^{-5} (Eq. 18) considering α = 80d cm^{-1} (Uslu *et al*., 2010); $\mu_{\beta l}$ = 0.288 p (Suriati *et al*., 2018); nozzle diameter 7.0× 10^{-4} and flow Q_γ = 600L h^{-1} (experimental device specifications); G_γ = 0.015 m^3 s^{-1} (Eq. 40). Nozzle flow (Q_γ = 600 L h^{-1}) is neglected with respect to G_γ (53280 L h^{-1}) of Eq. (40).

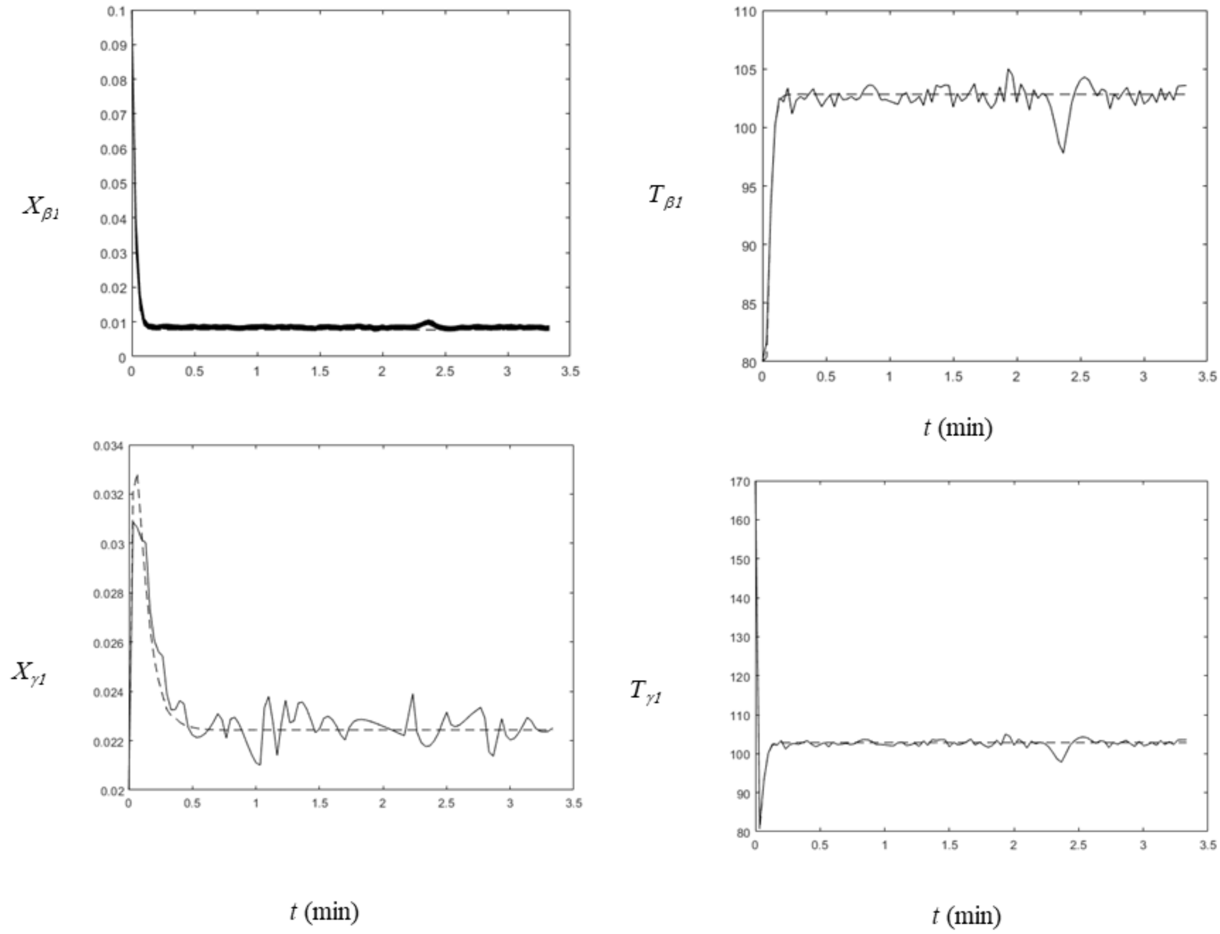

Fig. 2. Aloe vera spray drying dynamic simulation with punctual heterogeneous (—) and homogeneous (- - -) model at run 8 conditions of Tables 1-3.

Examples of spray drying simulation at two conditions (from Table 1) are plotted in Figs. 1 and 2. Continuous lines represent punctual heterogeneous model results and discontinuous lines the homogeneous one. The most notable fact observed in Figs. 1-2 is the chaotic behavior of punctual heterogeneous model and deterministic behavior of homogeneous model. In a phase diagram, the 22 state variables produce a periodic strange attractor. This chaotic behavior was reported for spray drying dynamic with lumped heterogeneous model by Palencia *et al*., (2002). However, the referred chaotic behavior is periodic around the homogeneous model steady state, except for dried product outlet moisture. The dried product outlet moisture and temperature is the second notable fact in Figs 1-2; it is evident that the homogeneous model assumption for dried solids output moisture ($X_\beta = X_{\beta i}$) is not applicable, as it was expected due to particle diameter ((6.5-7.0)$\times\ 10^{-5}$) predicted by Eq. (18). However the same assumption for temperature ($T_\beta = T_i$) is valid as can be observed in Figs. 1-2 for $T_{\beta 1}$ in which are plotted the 10 $T_{\beta j}$ (the 10 continuous lines are overlapped) resulting from Eq. (16). Therefore, the homogeneous model can approximate the air outlet conditions without the chaotic behavior of heterogeneous model but the outlet dried product moisture must be estimated with the averaged behavior of heterogeneous model.

The averaged (over particle volume and time) outlet solids particles moistures obtained by simulation at pseudo steady state, for the entire Table 1 treatments, are listed in Table 3, joint with the experimental moistures. It is important to remark that the simulated averaged moistures are pure mathematical estimation. There are many physical properties that cannot be experimentally obtained in this research. As example, particle diameter (it was estimated with Eq. 18), heat and moisture conductivity and diffusivity in particles (it was assumed equal to the reported by Aguirre-Alonso *et al*., 2019); heat and mass transfer coefficients (it was estimated with Eqs. 20-23). Nevertheless, the averaged simulated moisture obtained were between 50-80% lower than experimental one and were in the same magnitude order to experimental ones. Moreover, the dried product is highly hygroscopic and absorb water from moist environmental just when the dried product container is open, which explain why experimental moisture were greater than simulated ones in the whole of treatments. Resuming, both homogeneous and punctual heterogeneous models represent the thermodynamic behavior of Aloe vera extracts spray drying.

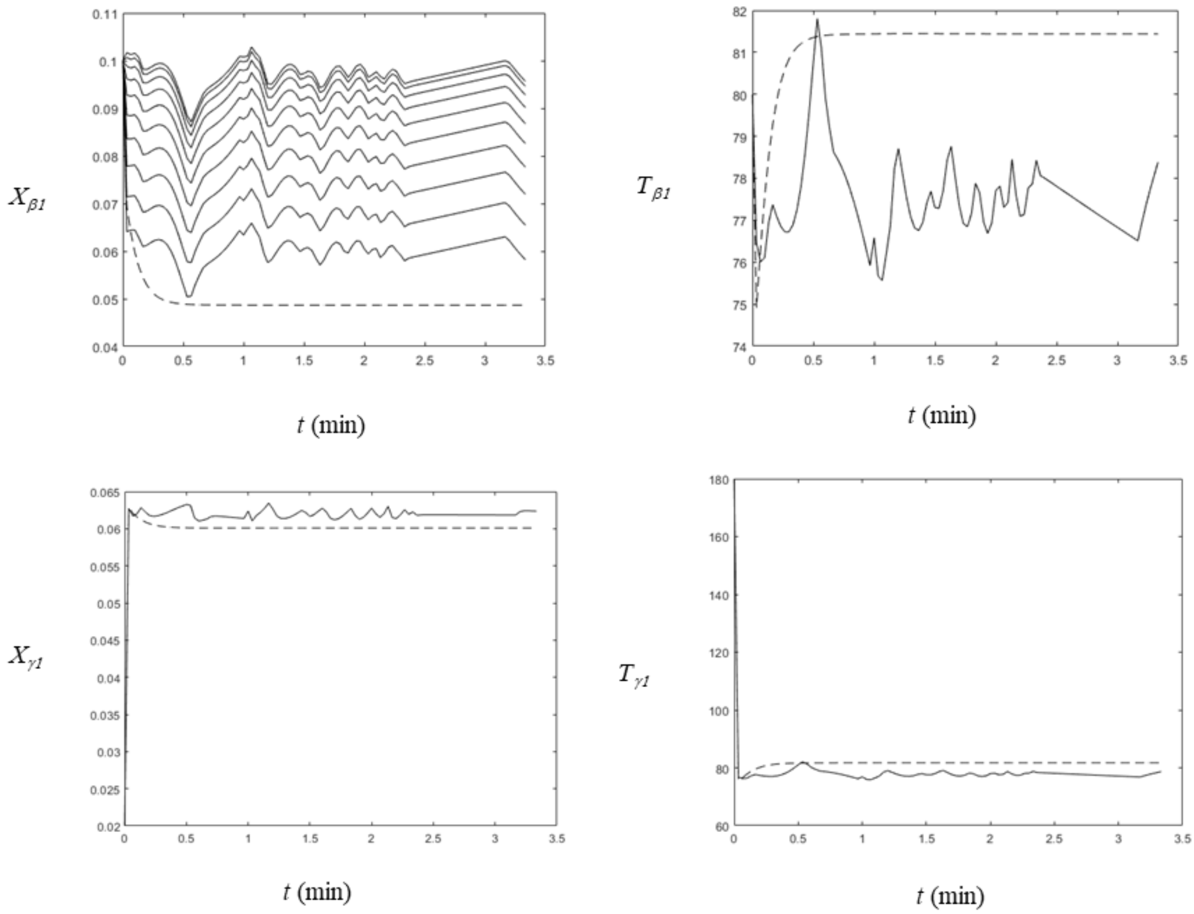

Fig. 3. Aloe vera spray drying dynamic simulation with punctual heterogeneous (—) and homogeneous (- - -) model at $T_{\gamma 1}$ =180ºC, $x_{\beta s}$ = 0.15, $Q_{\beta l}$=2 L h^{-1} and Table 2 variables.

4.3 Application to increase energy efficiency

It was written in introduction section that the rigorous thermodynamic modeling of spray drying process has the objective of to increase thermal efficiency. First law thermal efficiency may be declared as the ratio of the energy applied in water evaporation, evaluated as the inlet-outlet air moisture difference times the water evaporation latent heat, over the thermal energy applied to drying air, evaluated as environmental-inlet air enthalpy difference (Aguirre-Alonso *et al*., 2019),

$$\eta = \frac{(X_{\gamma 1} - X_{\gamma 0})\lambda}{h_{\gamma 0} - h_{\gamma env}} 100 \tag{43}$$

Applying Eq. (43) to Tables 1-3 variables and properties to homogeneous simulations (due to its deterministic nature) the results in first law thermal efficiency listed in Table 3 were obtained. The referred efficiencies vary between 3.7-16.0% which may be considered as poor efficiencies. These low thermal efficiencies are due to imposed constrains by the laboratory scale spray drier: nozzle neighborhood to dryer chamber wall leads moisture particles stick to wall and nozzle design is not proper for concentrated Aloe vera extracts. Aloe vera extract can be concentrated by evaporation until 15% of solids and it is important to remark that evaporation operation is more efficient than drying because is not limited by water-air equilibrium moisture (Eqs. 11-13). Then, assuming than a proper nozzle and a proper drying chamber (designed with CFD models) will be used with drying air inlet temperature of 180 ºC and well isolated drying chamber (U_{env} = 0), homogeneous and punctual heterogeneous models were used to simulate Aloe vera extracts spray drying with $x_{\beta s}$ = 0.15, $T_{\gamma 1}$ = 180ºC, G_{γ} = 0.015m^{3}s^{-1}; $Q_{\beta l}$ = 2Lh^{-1} and the variables and properties listed in Table 2. It was considering the same surface tension and $\mu_{\beta l}$ = 0.7p (resulting in 2R = 8.8× 10^{-5} m by Eq. 27) for the concentrated Aloe vera extract. Spray drying dynamic obtained at such conditions are plotted in Fig. 3 and the first law thermal efficiency reached 58.7 % (Eq. 43) with an averaged dried product moisture of 8.5% in dry basis (Fig. 3). As it was expected for grater particles the moisture profile within particles are more developed. Greater thermal efficiencies can be reaches by applying air recirculation or heat pump devices (Aguirre-Alonso *et al*., 2019). The important fact is that the proposed homogeneous-heterogeneous modeling can be used for thermal optimization of Aloe vera spray drying process.

Finally, the thermodynamically rigorous model proposed in present research can be combined with quality modeling, usually in terms of surface response models

(Rodrigues-Jimenes *et al.*, 2014) in order to propose spray drying thermal optimization problems with quality constrain.

Conclusions

Research hypothesis, that is, Eqs. (1)-(8), (11)-(15) and (18)-(29) represent a thermodynamically rigorous model for Aloe vera extracts spray drying process which was theoretically and experimentally validated. It is important to note that Eqs. (16)-(17) (discretized as Eqs. 26-29) solved until pseudo steady state, represent Fourier's and Fick's laws within spray dried particles with a continuous replace of energy and moisture due to a continuous particle inlet-outlet. Eqs. (16)-(17) can be solved only until a pseudo steady state because they manifested periodic chaotic behavior around averaged values. Homogeneous model solved until steady state result in deterministic values that approximate the homogeneous one and therefore it is a useful complement tool. The simulated averaged dried product moistures obtained with heterogeneous model were in the same magnitude order of experimental moistures obtained in a laboratory spray drier. The experimental assays first law thermal efficiencies were between 3.7-16.0%. A simulation showed that first law thermal efficiency can reach 58.7%. Therefore, proposed models can be solved jointly in order to solve energy efficiency maximization problems. It is feasible the combination of proposed models with quality models during spray drying processes in order to introduce quality constrains in the energy efficiency maximization problems.

Acknowledgment

Authors express their acknowledgements to Mexican Consejo Nacional de Ciencia y Tecnología (CONACyT) by scholarships of PhD student Cortés-Avendaño, E.J. (No. CVU 735363).

Nomenclature

a	Specific surface m^2m^{-3}
a	Thermodynamic activity
A	Transfer surface m^2
Cp	Constant pressure specific heat $Jkg^{-1}K^{-1}$
E	Total energy requirement $J\ kg^{-1}$
G	Mass flow $kg\ s^{-1}$
h	Enthalpy $J\ kg^{-1}$
j	Counter
k	Heat conductivity $J\ s^{-1}m^{-1}K^{-1}$
k_c	Mass transfer coefficient ms^{-1}
k_1, k_2, k_3	Sorption isotherms parameters
m	Mass g
M	Molecular weight g $gmol^{-1}$
N	Mass flux $kg\ m^{-2}s^{-1}$
p	Pressure Pa
q	Heat flux $Jm^{-2}s^{-1}$
Q	Volumetric flow Lh^{-1}
R	Particle radius m
r	Spherical radial coordinate m
t	Time s
T	Temperature K, ºC
U	Heat transfer coefficient $Js^{-1}m^{-2}K^{-1}$
V	Volume m^3
v	Velocity ms^{-1}
x	Mass fraction (or wet basis) kg kg^{-1}
X	Moisture mass relation (or dry basis) $kgkg^{-1}$
Greek symbols	
α	Surface tension d cm^{-1}
ε	Gas volume fraction
θ	Geometric factor
μ	Viscosity Pa s
η	Energy efficiency
λ	Evaporation latent heat J kg^{-1}
Ω	Topological factor
ρ	Density kg m^{-3}
Subscripts	
0	At drying chamber inlet
1	In drying chamber or at outlet
a	For air
env	At environmental conditions
exp	Experimentally obtained
l	For liquid
sim	Simulated
s	For solids
v	For vapor
w	For water
β	Particle phase
γ	Gas phase
Superscripts	
0	At reference
Dimensionless groups	
Nu	Nusselt number
Pr	Prandtl number
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number

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