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MODELING AND SELF-ORGANIZATION DYNAMICS OF AGGREGATION PROCESSES IN ACOUSTICALLY LEVITATED DISACCHARIDES SOLUTIONS

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Abstract. The present work deals with an analytical model formulated in order to characterize the drying process of acoustically levitated droplets. The model for the drying process is based on the calculation of the mass flux of vapor leaving the droplet surface and on the calculation of the change rate of the liquid droplet mass. The results obtained through the model, in terms of droplet diameter versus time, are compared with experimental data collected on aqueous solutions of trehalose and sucrose by using a video camera. It will be shown how the model predictions are in excellent agreement with the collected experimental data. In particular it emerges that the behaviour of squared droplet diameter as a function of time follows a linear trend for about 20 minutes, reaching then a plateau at longer time. Such a behavior can be explained in terms of water evaporation and disaccharide aggregative processes.

1. Introduction

Processes of solutions drying and self-organization dynamics of aggregation are enough complex mechanisms (Brenn *et al.* 1997; Yarin *et al.* 1998; Brenn *et al.* 2001; Groenewold *et al.* 2002; Yarin *et al.* 2002). In fact, although many theoretical models have been formulated, there are many few experimental data. Previous formulations furnish only integral formulations and no specific information on the single processes that occur. In this context, acoustic levitation is an instrument suitable for the study of droplets kinetics and it is fundamental to implement an intuitive model for drying processes and self-organization dynamics aggregation occurring when a single droplet is suspended in the air (Weber *et al.* 2013; Caccamo *et al.* 2017; Weber *et al.* 2017; Caccamo and Cannuli 2018; Cannuli *et al.* 2018). The analyses on a droplet during the evaporation process assent researchers to monitor all the physical and morphological changes. Also in biophysics field, aqueous solutions disaccharides (Magazù *et al.* 1989; Jannelli *et al.* 1996; Magazù 1996; Lokotosh *et al.* 2000; Magazù *et al.* 2008c, 2016), proteins (Magazù *et al.* 2010c, 2011b; Barreca *et al.* 2013; Fenimore *et al.* 2013), polymers (Aliotta *et al.* 1991; Branca *et al.* 1998, 1999a; Faraone *et al.* 1999a,b,c; Branca *et al.* 2000a,b; Magazù 2000; Triolo *et al.* 2000; Branca *et al.* 2002a,b; Migliardo *et al.* 2013b; Caccamo and Magazù 2016, 2017b,c; Caccamo *et al.* 2018b) and polyols (Magazù *et al.* 2007a), can be analyzed by the acoustic levitation technique, in particular for the study of hydrogen-bonded phenomena.

In this paper, the analyzed samples are diluted aqueous solutions of two disaccharides, i.e. trehalose and sucrose, in a concentration range of 50% water and 50% pure sample. So far a significant experimental material has been accumulated on the properties of disaccharides aqueous solutions, obtained by inelastic light diffusion (Magazù *et al.* 2013b) and incoherent neutron dispersion (Magazù *et al.* 2008a,b, 2011a).

An analytical model formulated in order to characterize the drying process of acoustically levitated disaccharide aqueous solutions is presented. The model for the drying process is based on the calculation of the mass flux of vapor leaving the droplet surface and on the calculation of the change rate of the liquid droplet mass. The results obtained through the model, in terms of droplet diameter versus time, are compared with experimental data collected on aqueous solutions of trehalose and sucrose by using a video camera, shown how the model predictions are in excellent agreement with the collected experimental data for the initial time trend. Finally, the study reports an analysis of the resulting aggregated structures evidencing the different homogeneity degree for the investigated systems.

2. Mathematical model for drying processes in acoustically levitated droplets

From a general point of view, the drying process of droplets is described by simple evaporation models based on the film theory, where it is assumed that the entire resistance to mass transfer is equal to the interface between the liquid and the gas phase. Figure 1 shows the concentration and temperature behaviors inside a gas film of thickness δ, in particular heat is transported only by diffusion towards the solution in exchange for mass diffusion from the drying solution in the opposite direction, so the gas film is surrounded by two boundaries: an initial boundary ($x = 0$) and an outer boundary ($x = \infty$) that represents the condition far away from the solution. The following assumptions are hypothesized:

- diffusion coefficient and thermal diffusivity are supposed constant and the flow is assumed laminar inside the film;
- temperature and concentration distributions depend only by the distance from the surface, at $x = 0$ from $T = T_{surface}$, $Y_c = Y_{surface}$ and at $x = \delta$ from $T = T_\infty$, $Y_c = Y_{c,\infty}$;
- the convection flows between the solution and the external environment are equal to the diffusion of the species and conduction of heat inside the film, arranging in the most appropriate way the values of δ_T and δ_C;
- no chemical reaction within the film occurs;
- thermo-physical properties are assumed to be spatially uniform;
- the thickness of the film (δ) is constant along the surface and it is independent of the mass transfer rate;
- the pressure drop does not influence the gas phase physical properties.

The mathematical model for drying processes in acoustically levitated droplets is based on the calculation of the mass flux of vapor leaving the droplet surface and on the calculation of the change rate of liquid droplet mass. The thickness thermal and diffusional films can be defined as:

$$\delta_c = \frac{2 \cdot r_s}{Sh - 2} \quad \text{and} \quad \delta_T = \frac{2 \cdot r_s}{Nu - 2} \tag{1}$$

with Sh and Nu, Sherwood and Nusselt numbers (Yarin *et al.* 1998), non-dimensional heat and mass transfer coefficients, defined as it follows:

$$Sh = \frac{D \left.\dfrac{\partial Y}{\partial r}\right|_s}{Y_\infty - Y_s} \quad \text{and} \quad Nu = \frac{D \left.\dfrac{\partial T}{\partial r}\right|_s}{T_\infty - T_s} \tag{2}$$

The liquid vapor flow rate at the droplet surface can be calculated as:

$$\dot{m}_v = \dot{m}_v Y_s - \pi D^2 \rho_{gas} D_g \left.\frac{\partial Y_{vap}}{\partial r}\right|_s \tag{3}$$

with Y_{vap}, mass fraction at the droplet surface, supposed to be uniform.

By substituting (2) in (1) it is possible to obtain:

$$\dot{m}_v = \pi D \rho_{vapor} D_g Sh \left(\frac{Y_{vap,s} - Y_{vap,\infty}}{1 - Y_{vap,\infty}} \right) \tag{4}$$

where B_M is the Spalding mass transfer coefficient:

$$B_M = \frac{Y_{vap,s} - Y_{vap,\infty}}{1 - Y_{vap,\infty}} \tag{5}$$

The basic phase equation for the conservation of mass can be expressed as:

$$\frac{d}{dr}\left(\rho_{gas} r^2 U(r)\right) = 0 \tag{6}$$

where U is the radial velocity of the gas surrounding the droplet.

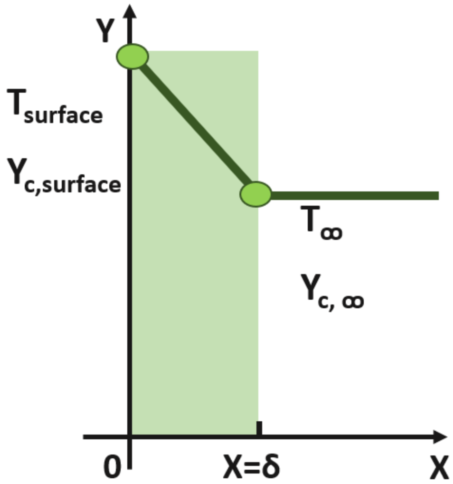


FIGURE 1. Concentration and temperature behaviors inside a gas film of thickness δ.

From the integration of (6) it is possible to obtain:

$$r^2\rho_{gas}U(r) = \frac{\dot{m}_v}{4\pi} = const \tag{7}$$

Imposing the conservation of the species vapor of the evaporating component:

$$\frac{d}{dr}\left(\rho_{gas}r^2U(r)Y_{vap}\right) = \frac{d}{dr}\left(\rho_{gas}r^2D_g\frac{dY_{vap}}{dr}\right) \tag{8}$$

and substituting (6) into (8), the resulting equation is:

$$-\frac{dY_{vap}}{1-Y_{vap}} = \frac{\dot{m}_v}{4\pi\rho_{gas}D_g}\frac{dr}{r^2} \tag{9}$$

From the integration of (9) along the film thickness δ with boundary conditions, i.e.:

$$Y_{vap} = Y_{vap,s} \quad \text{at} \quad r = r_s \quad \text{and} \quad Y_{vap} = Y_{vap,\infty} \quad \text{at} \quad r = r_{f,M} \tag{10}$$

it results:

$$\ln\left(\frac{1-Y_{vap,\infty}}{1-Y_{vap,s}}\right) = -\frac{\dot{m}_v}{4\pi\rho_{gas}D_g}\left(\frac{1}{r_{f,M}} - \frac{1}{r_s}\right) \tag{11}$$

Re-arranging (11) for $\dot{m}_v$:

$$\dot{m}_v = 4\pi r_s^2\rho_{gas}D_g\ln\left(1+B_M\right)\cdot\frac{r_{f,M}}{R_s\left(r_{f,M}-r_s\right)} \tag{12}$$

and hence:

$$\dot{m}_v = \pi D\rho_{gas}D_g\ln\left(1+B_M\right)\cdot\frac{r_{f,M}}{r_s\left(r_{f,M}-r_s\right)} \tag{13}$$

Drying processes imply a time in which the drying rate is constant and the solution evaporation rate from the droplet surface has the same behavior of a pure liquid of the same size. It follows the D^2-law, that predicts that the square of the droplet diameter decreases linearly with time, followed by a period in which the rate of evaporation falls. Few important assumptions must be considered for the levitated droplet, such as:

- heat and mass transfer between liquid and gas phase are diffusion controlled;
- a spherical symmetry is considered;
- no radiation effects take place;
- constant and uniform temperature occurs.

The change rate of the liquid droplet mass is expressed as it follows:

$$\dot{m}_d = -\frac{dm_d}{dt} \tag{14}$$

$$\frac{dm_d}{dt} = -\rho_{droplet}\frac{dV}{dt} = -\rho_{droplet}\frac{\pi}{6}\frac{dD_s^3}{dt} \tag{15}$$

Being $\dot{m}_v = -\dot{m}_d$, then comparing eqn. (15) with the (13) the equation for the diameter becomes:

$$\pi D\rho_{vapor}D_gSh\left(\frac{Y_{vap,s}-Y_{vap,\infty}}{1-Y_{vap,\infty}}\right) = -\rho_{droplet}\frac{\pi D}{6}\frac{dD_s^2}{dt} \tag{16}$$

with $D_S^3 = D_S^2 \cdot D$; in other terms D_S^3 can be partitioned into a constant diameter value, i.e. D, and in a squared diameter value variable as function of time, D_S^2.

$$\frac{dD_s^2}{dt} = -6\frac{\rho_{gas}}{\rho_{droplet}} D_g Sh \left(\frac{Y_{vap,s} - Y_{vap,\infty}}{1 - Y_{vap,\infty}} \right) = -6\frac{\rho_{gas}}{\rho_{droplet}} D_g Sh \ln(1 + B_M) \tag{17}$$

For a diffusion controlled evaporation process, the film thickness goes into infinity. The modified Sherwood number Sh takes the value 2 and after substituting the value of Sh into (15), (17) becomes:

$$\frac{dD_s^2}{dt} = -12\frac{\rho_{gas}}{\rho_{droplet}} D_g \ln(1 + B_M) \tag{18}$$

$$\int \frac{dD_s^2}{dt} = \int -12\frac{\rho_{gas}}{\rho_{droplet}} D_g \ln(1 + B_M) \tag{19}$$

The integration of this equation for an initial condition D_0, i.e. at $t = 0$, gives the D^2-Law that describes the temporal evolution of droplet surface of pure liquid:

$$D_s^2 = D_{0s}^2 - \beta t \tag{20}$$

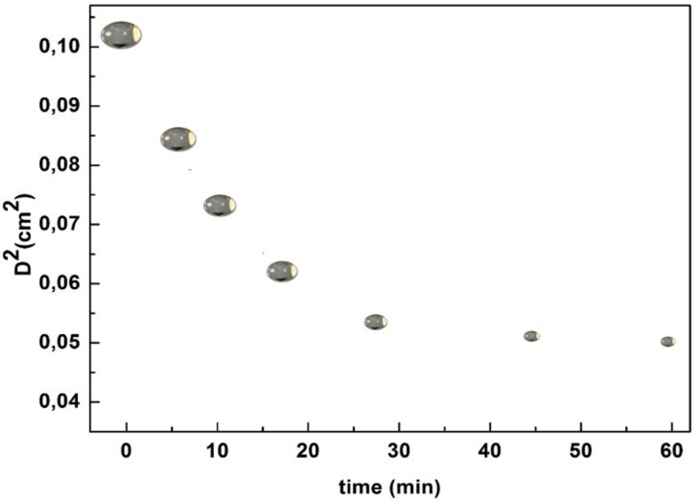
where β is the evaporation rate coefficient:

$$\beta = 12\frac{\rho_{gas}}{\rho_{droplet}} D_g \ln(1 + B_M) \tag{21}$$

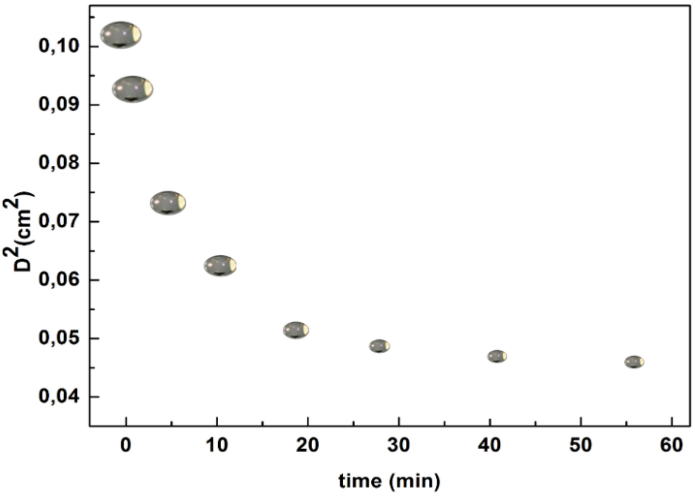
The density of the liquid droplet is $\rho_{droplet}$, the density of the gas at the droplet surface is ρ_{gas}, the diffusion coefficient is D_g. In addition to the evaporation rate, another important parameter in droplet evaporation is the lifetime of the droplet, also called evaporation time τ_{end}, which can be determined from (18) with $D|_{\tau=\tau_{end}} = 0$: $\tau_{end} = \dfrac{D_0^2}{\beta}$.

3. Results and discussion

The mathematical model for drying processes in acoustically levitated trehalose and sucrose aqueous droplets, manufactured by Sigma-Aldrich, as a function of disaccharide weight fraction percentage is tested by a direct measurement performed by means of a Hitachi KP-HD20A video camera, with a resolution of 1944 H x 1092 V (2.1 megapixels) (Magazù *et al.* 1998a,b; Branca *et al.* 1999b,c; Magazù *et al.* 1999a,b; Ballone *et al.* 2000; Branca *et al.* 2002c, 2003, 2005; Magazù *et al.* 2007b; Minutoli *et al.* 2007, 2008; Pagnotta *et al.* 2008; Varga *et al.* 2008; Barreca *et al.* 2010; Magazù *et al.* 2010a,b; Condello *et al.* 2012; Magazù *et al.* 2013a,c; Migliardo *et al.* 2013a, 2014). Double-distilled water was used in making the solutions. Measurements for each solution were repeated four times. Droplets of 1.5 mm of radius were inserted into the pressure node of the sound field using a micropipette injection. Figure 2 reports the D^2 data of trehalose and sucrose aqueous solutions as a function of time. As it can be seen, for both trehalose and sucrose aqueous solutions, D^2 vs t follows a linear trend for about 20 minutes, reaching then a plateau value at longer time. This result shows how both samples are able to avoid total water evaporation since a percentage of water is strongly bonded to the disaccharide. Such a behavior can be explained in terms of disaccharide aggregative processes. From the diameter decay law, it also emerges that water molecules are bonded to trehalose with a higher strength in respect to sucrose. In a future work levitation technique will be employed in conjunction



(a)



(b)

FIGURE 2. D^2 data of the trehalose (on the top) and sucrose (on the bottom) aqueous solution droplets as a function of time, performed by a Hitachi KP-HD20A video camera, with a resolution of 1944 H x 1092 V (2.1 megapixels).

with spectroscopic techniques to validate the analytical model and wavelet analysis will be employed for removing noise from the spectra (Magazù *et al.* 2012; Caccamo *et al.* 2016; Caccamo and Magazù 2017a; Marchese *et al.* 2017; Caccamo *et al.* 2018a).

4. Conclusions

In this paper, an analytical model has been formulated in order to characterize the drying process of acoustically levitated droplets of trehalose and sucrose. The results obtained through the formulated approach are compared with experimental data collected by using Hitachi KP-HD20A video camera. From the collected data it emerges that the behaviour of D^2 vs t follows a linear trend for about 120 seconds, reaching then a plateau value at longer times. It is shown how the model predictions are in excellent agreement with the collected experimental data. It clearly emerges that both the disaccharides are able to avoid total water evaporation. From the diameter decay law, it also emerges that water molecules are more strongly bonded to trehalose in respect to sucrose.

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