

Theoretical and Experimental Investigation of Particle Formation from Evaporating Microdroplets



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Introduction

The present study introduces a new approach for the design of structured microparticles from evaporating solution and suspension microdroplets. A model is proposed for calculating the transient concentration profiles of solutes during the evaporation process until the onset of shell formation. The model can predict various properties of the final dry particle. The model is an extension of the classical VFL particle formation model [1-3]. A novel monodisperse droplet chain setup was used to verify the model results by generating an evaporating, widely spaced droplet chain in a laminar gas flow with controlled temperature and relative humidity. The generated droplet chain also allowed for the determination of the droplet evaporation rate from the droplet position and velocity.

Method and Materials

i- A newly developed normalization of the diffusion equation in one-dimensional spherical coordinates allows the determination of the transient concentration profiles in terms of non-dimensional process parameters [4-5].

$$\frac{\partial c}{\partial \tau} = \frac{1}{2Pe(1-\tau)} \left[\frac{\partial^2 c}{\partial R^2} + \frac{2}{R} \frac{\partial c}{\partial R} \right] - \frac{R}{2(1-\tau)} \frac{\partial c}{\partial R}$$

ii- Various dry particle properties can be calculated based on the model results [4-5].

$$d_v = \sqrt{1 - \tau_{sat}} d_o \quad \text{Volume equivalent diameter}$$

$$\rho_p = C_o \left[\frac{d_o}{d_v} \right]^3 \quad \text{Particle density}$$

$$d_a = \sqrt{\frac{\rho_p C_o (d_o)^3}{\rho C_o (d_a)^3} \frac{1}{\lambda}} d_v \quad \text{Aerodynamic diameter}$$

$$\frac{d_{sh}}{d_o} = \sqrt[3]{1 - \frac{\rho_p}{\rho_{sh}}} \quad \text{Shell thickness}$$

iii- The droplet evaporation rate can be calculated by solving the droplet equation of motion and measuring its trajectory data.

$$F_D + F_G = \frac{d(mv)}{dt}$$

iv- The droplet chain setup was used to validate the model results and to obtain the droplet trajectory data during evaporation.

c	Normalized concentration
R	Normalized radial coordinate
$\tau_{sat} = 1 - \tau_{in}$	Precipitation time
τ_{sat}	Saturation time
C_o	Slip correction factor
d_o	Initial droplet diameter
d_a	Internal particle diameter
ρ_{sh}	Shell density
λ	Mean free path of the drying gas
Pe	Peclet number

Results

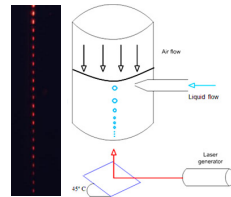


Fig.(1) Stable and well controlled droplet trajectories allow determination of evaporation rates. $d_o = 85 \mu\text{m}$, $T_{air} = 90^\circ\text{C}$, $f = 300 \text{ Hz}$

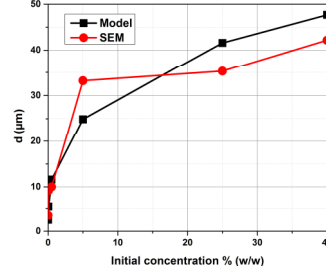
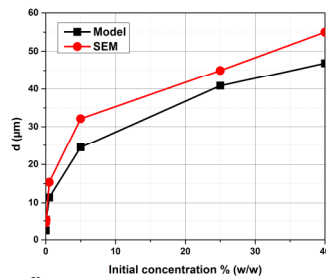


Fig.(2) Diameter of the dry particle formed at 85 °C (top) and 125 °C (bottom) at different initial concentrations. Model results vs. SEM images.

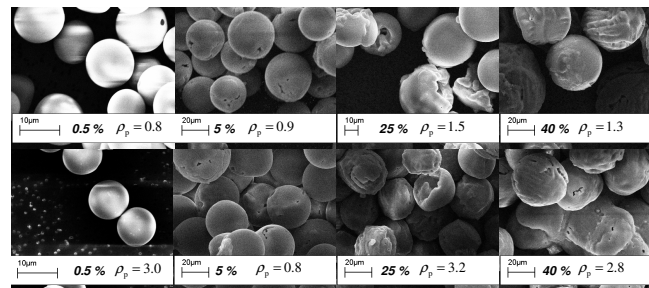


Fig.(5) SEM images of sodium nitrate particles formed at 85 °C (top) and 125 °C (bottom) at different concentrations. As the initial concentration increases the precipitation time increases which promotes crystallization. Particle morphology change from smooth to wrinkled with the increase in concentration. Particle density is in g/cm^3 .

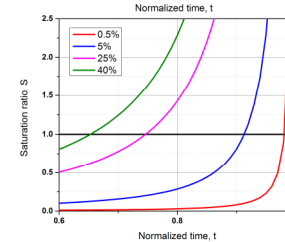
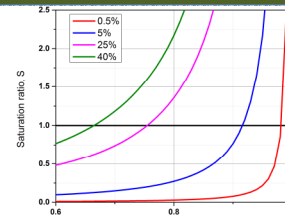


Fig.(4) Sodium nitrate surface saturation at 85 °C (top) and 125 °C (bottom). The time axis shows the time needed to reach saturation (i.e. $s=1$) as fraction of the total droplet drying time. As the initial concentration increases, the time needed to reach saturation decreases with a subsequent increase in the precipitation time.

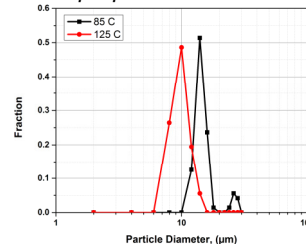


Fig.(3) Dry particle diameter count distribution for particles formed from 0.5% solution droplets showing the narrow distribution of the particles formed using the droplet chain technique.

Discussion and Conclusion

The proposed approach has many advantages as evident from the results.

1- The droplet chain setup is a good technique in generating a stable droplet chain with a known initial diameter in a controlled conditions which allows the calculation of the droplet evaporation rate (Fig.(1)).

2- The droplet chain and consequently the final dry particles have a narrow size distributions which can be used to verify the model results (Fig.(2) & Fig.(3)).

3- The proposed model can predict the time needed to reach saturation on the droplet surface (Fig.(4)) which is used to calculate the dry particle size (Fig.(2)) and the precipitation time left for the crystallization process.

4- The change of the particles morphology (Fig.(5)) from smooth to wrinkled particles correlate with an increase of the precipitation time as predicted by the model.

The present approach advantages can be summarized as following

- Easy determination of the evaporation rate
- Fast calculation of the final dry particle properties.
- Well suited for the design of structured, multi-layered and multi-component microparticles.
- The ability to correlate the dry particle properties to different process parameters.

References

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