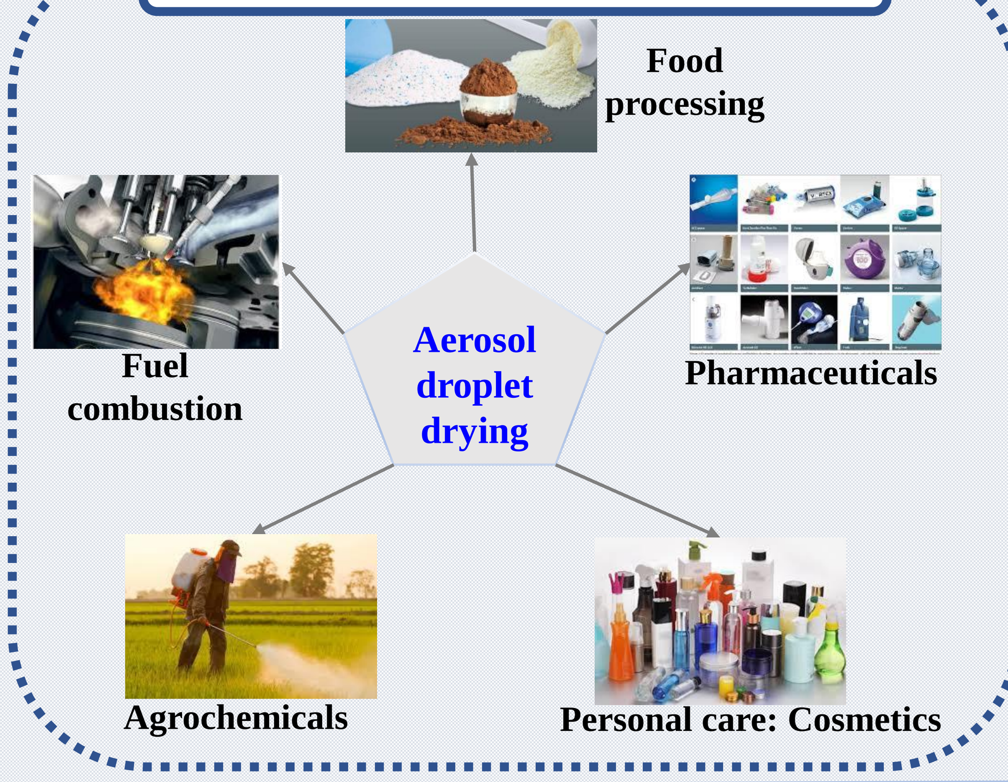
SINGLE DROPLET DRYING KINETICS AND PARTICLE FORMATION FROM AEROCOLLOIDAL SUSPENSION MICRODROPLETS

University of Bristol
Bristol Aerosol Research Centre

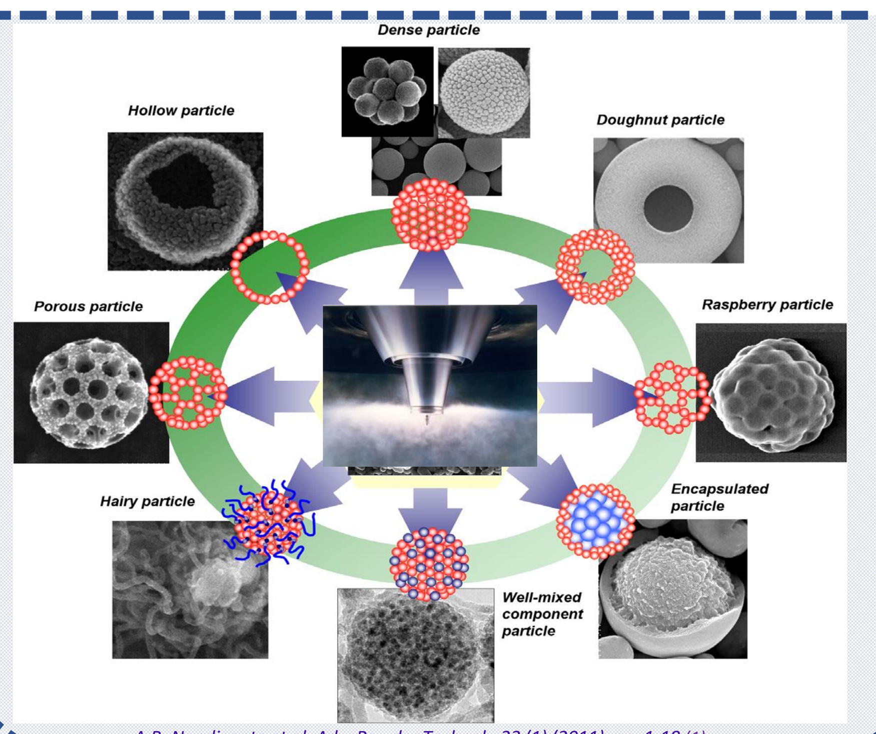
Justice Archer*, Jim S. Walker, Florence K. A. Gregson, Dan A. Hardy and Jonathan P. Reid
University of Bristol (Email: justice.archer@bristol.ac.uk)

EPSRC Engineering and Physical Sciences Research Council

Why Study Aerosol Droplet Drying?



Morphologies of Dried Aerosol Microparticles



A.B. Nandiyanto et al, Adv. Powder Technol., 22 (1) (2011), pp. 1-19 (1)

Péclet number: Describing Dry Aerosol Particle Morphology

$$Pe_i = \frac{\kappa}{8D_i}$$

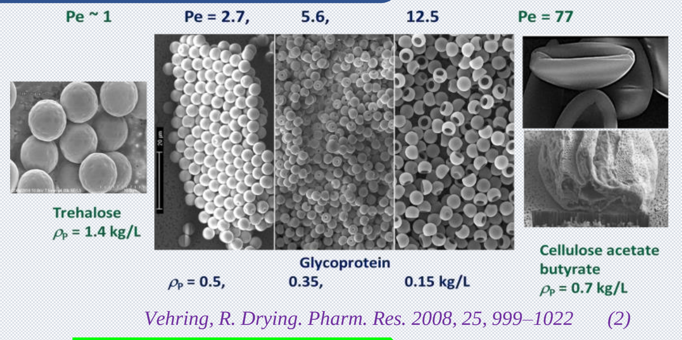
κ: Evaporation rate
Rate of droplet surface recession via evaporation.

$$\kappa = -\frac{dd^2}{dt}$$

D_i: Solute diffusion constant
Diffusion of suspended inclusions within droplet to the droplet surface

$$D_i = \frac{KT}{6\pi\mu R_i}$$

If $Pe << 1$ → homogeneous mixing of inclusions within droplet - *dense particle formulation*
$Pe >> 1$ → surface enrichment of inclusions at the droplet surface - *hollow or buckled particle formation*

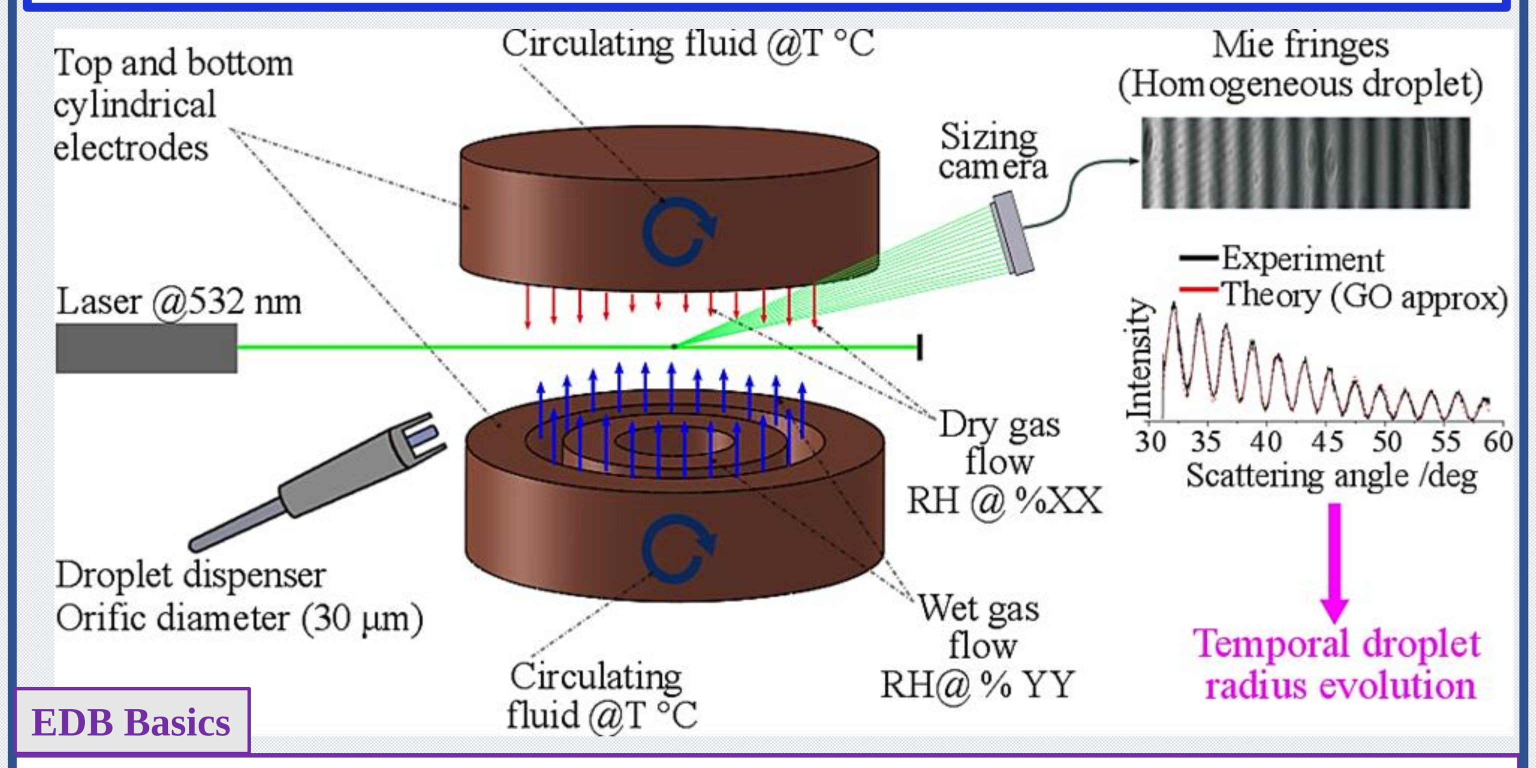


Vehring, R. Drying. Pharm. Res. 2008, 25, 999–1022 (2)

Purpose

- To provide comprehensive experimental study for improved understanding of the influence of evaporative and inclusion parameters on droplet drying kinetics and morphology of final dry products, particularly those encountered in spray-drying processes.

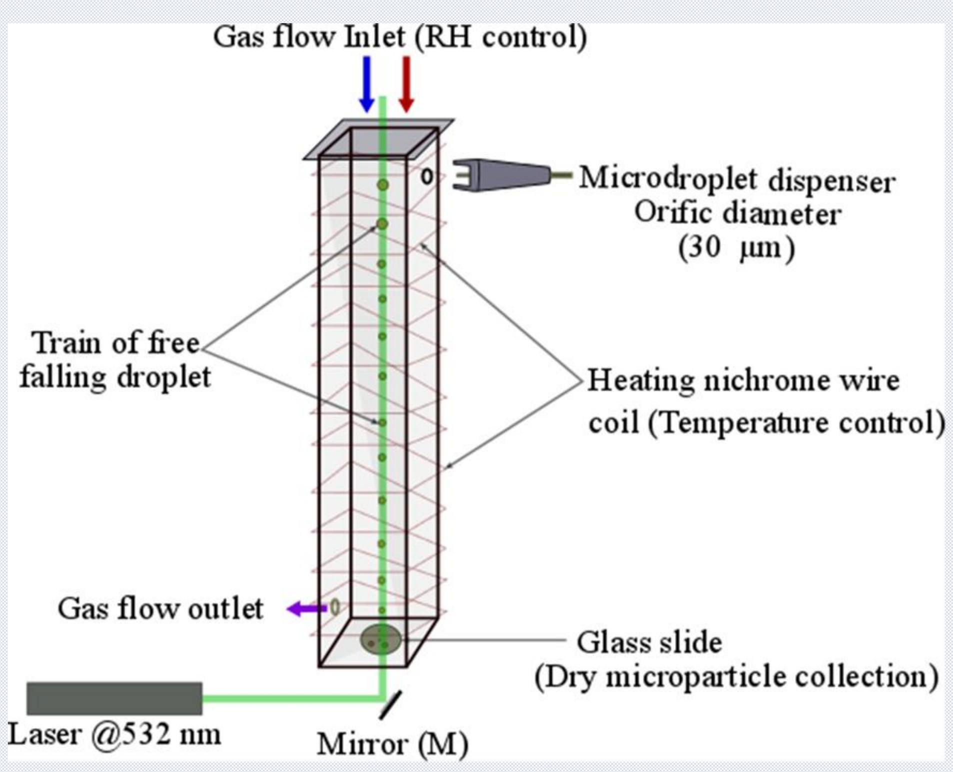
Electrodynamic Balance (EDB) for Droplet Drying Kinetic Measurement



EDB Basics

- An AC fields is applied to the top and bottom electrodes to produce a time-varying electric (AC) fields that confines a charged droplet in *"free space"* at the centre of the trap.
- Gravitational, drag force from gas flow and electrostatic force are balanced by static DC voltage applied between the electrodes.
- Single droplets constrained at the trap centre is illuminated with a 532 nm CW laser, and the resulting elastically scattered laser light from the spherical, liquid droplet in the form of light and dark fringes is collected over an angular range ~24° centred at 45° to the forward direction of the laser. The fringe spacing in the elastic light scattering, relates to the droplet size.

The Falling Droplet Column (FDC): *for Collecting Dried Microparticles and Offline Imaging Analysis*

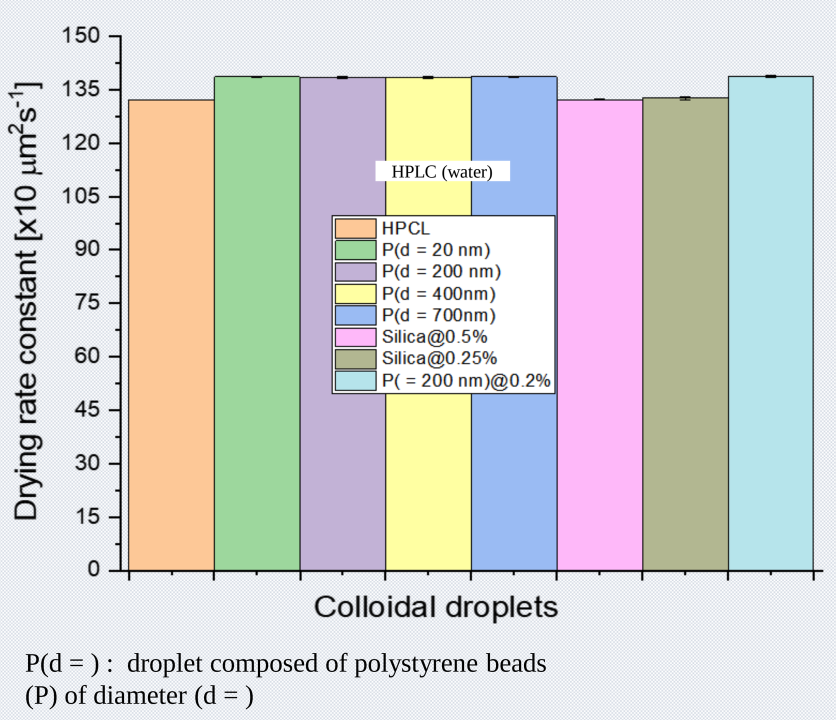


A chain of droplet dispensed by the microdroplet dispenser propagates downwards through the glass column.

The droplets evaporate as they fall down the column, so that dried particles are collected at the bottom. The final dried particle surface properties and morphologies are analysed offline using Scanning Electron Microscopy (SEM).

With the same drying conditions in the FDC and the EDB, the aerosol **droplet drying kinetics** can be correlated directly to the **final properties of the dried particles**

Effect of Initial Droplet Composition on Aerosol Droplet Evaporation Rates



P(d =) : droplet composed of polystyrene beads
(P) of diameter (d =)

The figure shows evaporation (drying) rates of different aerocolloidal droplets (hydrophilic silica and polystyrene beads) compared with the evaporation rate of water (HPCL)

The evaporation rate were obtained by fitting a straight line to the **first stage** of the evaporation process where the temporal evolution of the **droplet surface area** follows isothermally as the well-known ***"R - squared law*** and are very similar to the rate constant for the pure water droplet.

Thus, the starting composition of the aerocolloidal droplets does not strongly influence the solvent evaporation/drying rate with the included particles acting as **spectators**

Acknowledgement

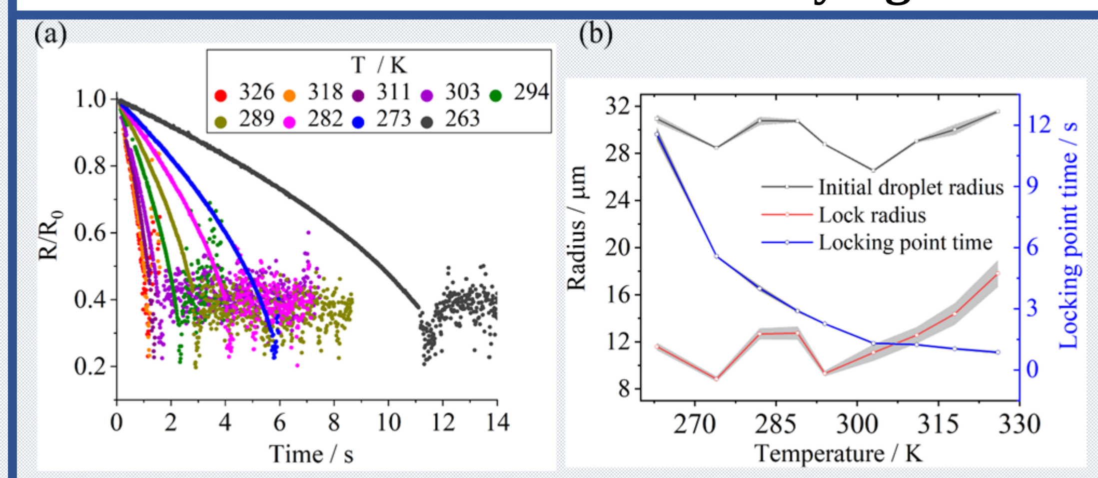
The authors thank Funding from EPSRC and support from Bristol Aerosol Research Centre for supporting this work

References

1. A.B. Nandiyanto, et. al, Adv. Powder Technol., 22 (1) (2011), pp. 1-19
2. R. Vehring, Pharm. Res. 2008, 25, 999–1022
3. L. Korson, et. al Phys. Chem. 1969, 73 (1), 34–39
4. Baldelli A. Boraey, et.al Molecular Pharmaceutics, **12** (8), 2562-2573, 2015

Results:

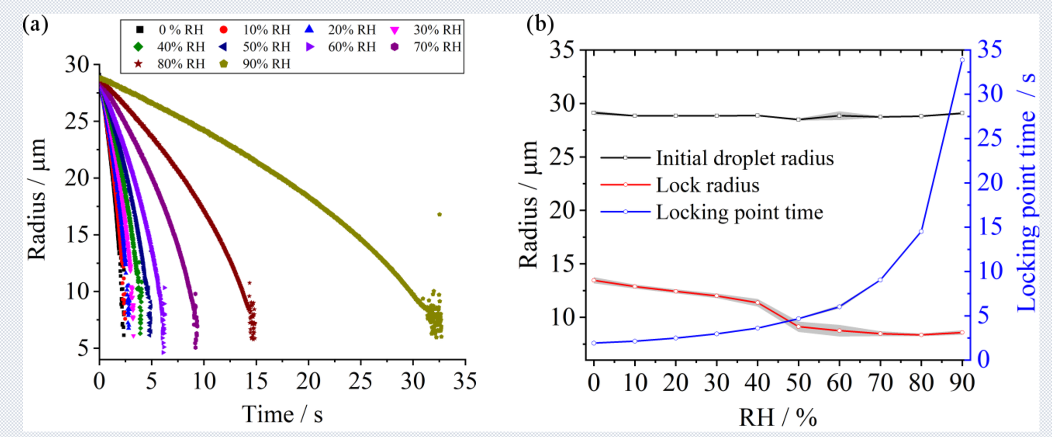
Influence of Gas Phase Drying Conditions on Aerosol Droplet Drying Kinetics



Influence of Temperature on Aerosol Droplet Drying Kinetics:

Evaporation profile of 0.6 v/v % colloidal silica droplets drying at varying temperatures. (a) normalized radius (R/R_0) as a function of time. The droplet evaporation rate increases with increasing gas phase temperature.

(b) Dependence of the inclusions surface locking points on temperature. The locking point time (LPT) and the radius at which the locking point time occurs (lock radius) is defined as the **onset** of the ***first visual skin or shell formation*** and morphology development where the ***surface properties*** of the drying droplet transitions from a *smooth liquid surface* to a ***rough solid phase***. For T = 263 K , the locking point time is **~ 11.3 s** with the lock radius at **~ 11.8 μm**

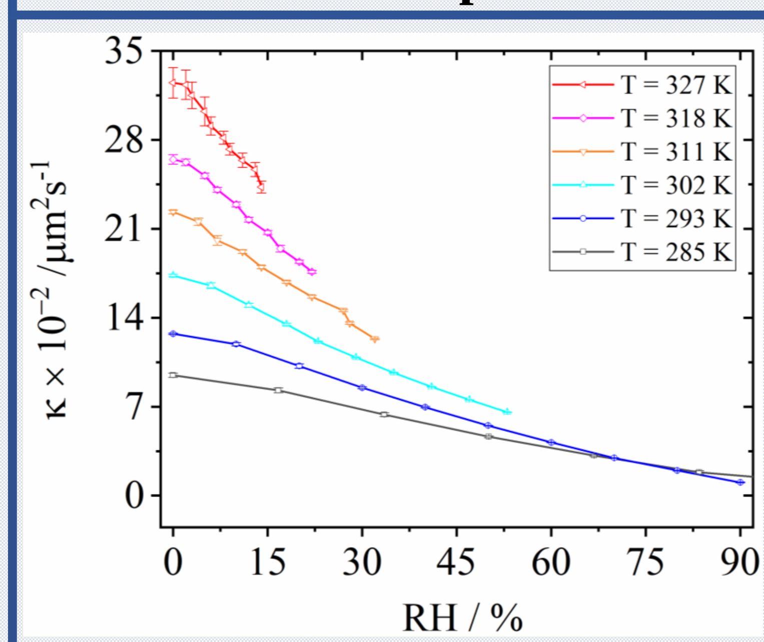


Influence of Relative Humidity on Aerosol Droplet Drying Kinetics:

(a) Evaporation profiles of 0.5 vol.% aerocolloidal silica droplets at 294 K over a range of RH from 0 - 90 %. The droplet evaporation rates increases with decreasing relative humidity.

(b) Initial droplet radius (dark line), lock radius (red line) at which the locking point time (blue line) occurred. The shaded region and the error bar are the standard deviation for over 15 droplets averaged for each droplet kinetic measurement.

Influence of Temperature and Relative Humidity on Evaporation Rate and Péclet Number

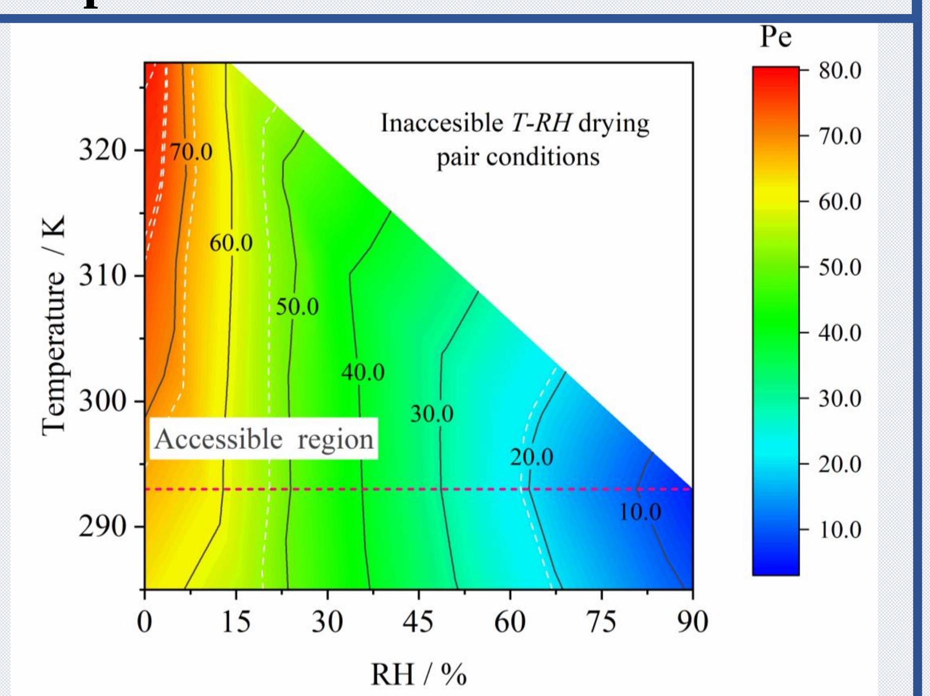


Evolution of evaporation rate (κ) from kinetic measurements for droplets drying at different gas phase temperatures into varying humid conditions. The error bars are standard deviations from evaporation rate constants obtained from averaging 20 droplets at each measurement.

$$Pe_i(T, RH, r_i) = \frac{3\pi\mu_i(T)r_i}{4KT}\kappa(T, RH)$$

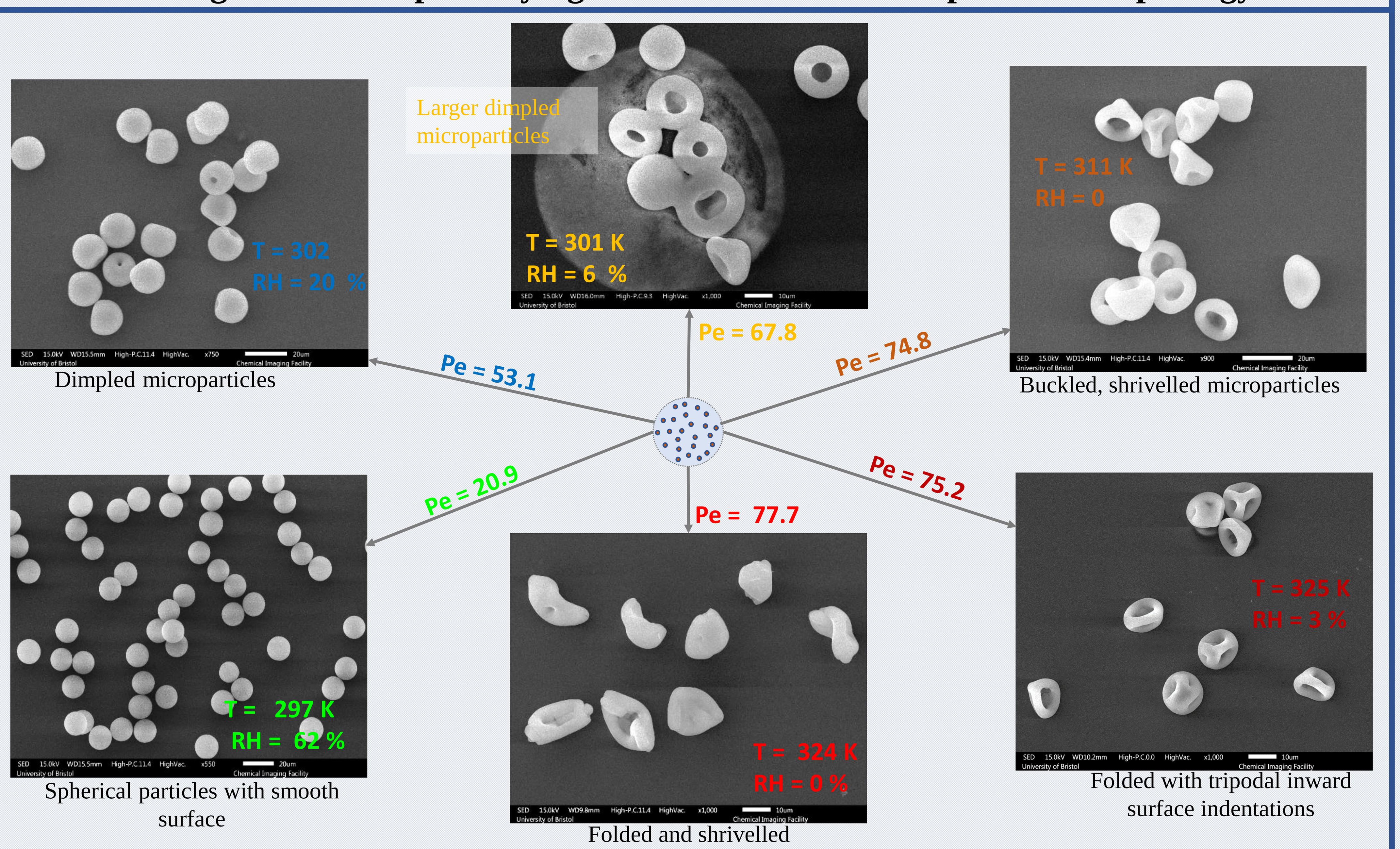
$$\log\frac{\mu_i(T)}{\mu_0(T_0)} = \frac{A(20-T)-B(T-20)^2}{C+T} \quad [3]$$

A, B, C are constants, $\mu_0(T_0)$ is the viscosity of water at T =293 K



Pe-T-RH Phase Map** for a* **0.4 % v/v** silica droplets evaporating at different drying temperatures into varying % RH conditions. The ***Pe number appears to be **independent** of temperature at higher RHs

Relating Aerosol Droplet Drying Kinetics to Dried Microparticle Morphology



SEM micrographs of dried silica microparticles from 0.4 % v/v colloidal silica droplets produced by a droplet chain technique at increasing Péclet numbers for temperature - RH drying condition pairs.

Conclusions

- ✓ **EDBs can be used to:** Measure ***aerosol droplet drying kinetics to obtain evaporation rates*** constants and to predict onset of inclusions surface shell/skin formation under different drying conditions from 263 -330 K and 0 - 90 % RH
- ✓ Priori knowledge of aerosol droplet drying kinetic measurements can be used to predict and relate final morphology of dry aerosol microparticles using the "*Pe-T-RH phase maps*".
- ✓ Aerocolloidal droplets of different initial compositions evaporating into similar drying conditions have comparable ***evaporation rates constants*** within **the steady-state diffusion regimes** with the included particles ***acting as spectators.***