Impact of humidity on silica nanoparticle agglomerate morphology and size distribution

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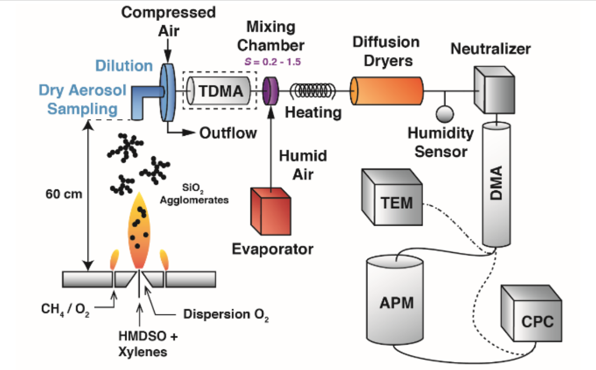
Motivation

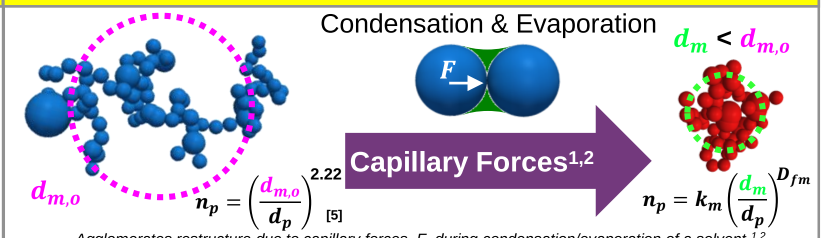
In the presence of humidity, agglomerates of single primary particles (PPs) and/or chemically-bonded ones (aggregates) restructure, forming smaller and more compact structures.[1,2] This is known to affect the fluidization[3] and spray drying[4] of soot. Here, the evolution of silica morphology and mobility size distribution processed under humid conditions is monitored for the first time and compared to humidified soot nanoparticles.

Processing of SiO_2 Agglomerates with Water Vapor



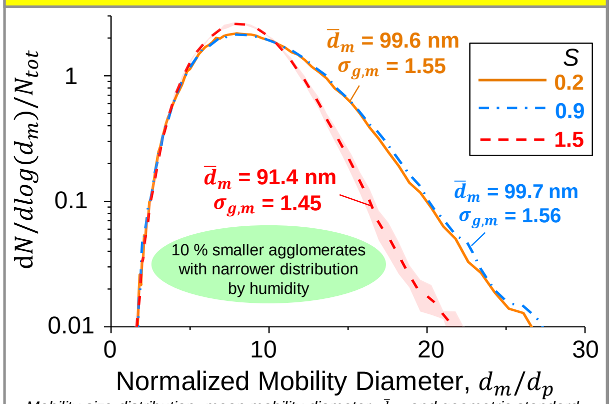
Silica agglomerates are sampled above the flame, diluted and mixed with humid air with varying temperature to achieve saturation ratios, S, from 0.2 to 1.5. The diluted sample is directed through two diffusion dryers to a differential mobility analyzer (DMA) and then to an aerosol particle mass analyzer (APM), a condensation particle counter (CPC) or collected for transmission electron microscopy (TEM). During tandem-DMA (TDMA) measurements, dry agglomerates with mobility diameter, $d_{m,o}$, are selected by TDMA before mixing with humid air.

Agglomerate Characterization by Scaling Laws



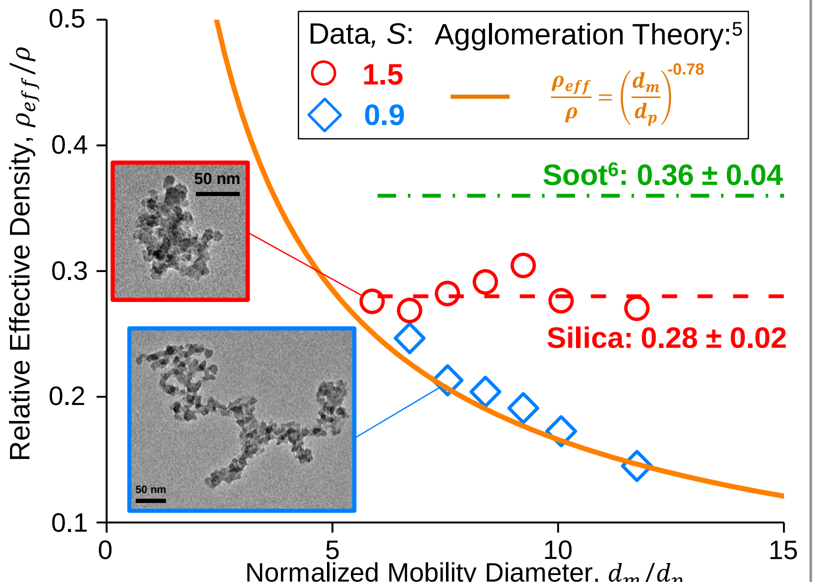
Agglomerates restructure due to capillary forces, F, during condensation/evaporation of a solvent.[1,2] (n_p, d_p: PP number and diameter; D_{fm}, k_m: mass-mobility exponent and prefactor)

Shrinking of Agglomerate Mobility Size



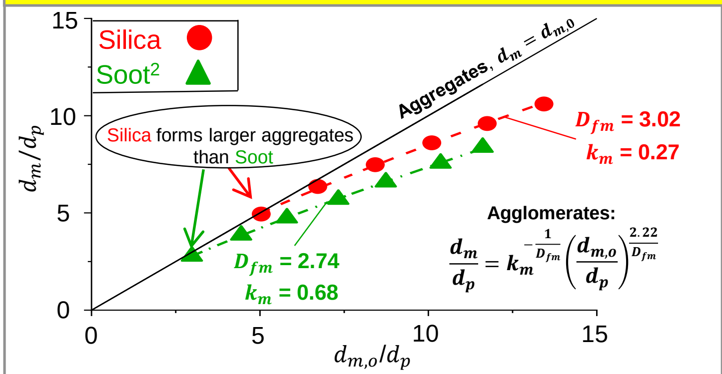
Mobility size distribution, mean mobility diameter, $\bar{d}_m$, and geometric standard deviation, $\sigma_{g,m}$, of silica agglomerates after exposure to different saturation ratio, S.

Drastic Change of Agglomerate Morphology by Humidity

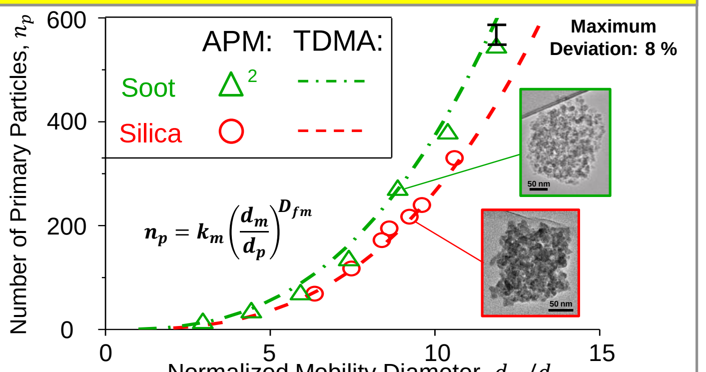


Relative effective density, ρ_{eff}/ρ, of silica agglomerates exposed to different S (symbols) compared to soot agglomerates[6] with smaller PP standard deviation, $\sigma_{g,p}$, (dot broken line) and agglomeration theory[5] (solid line).

Mass-mobility Relationship of Restructured Agglomerates measured by TDMA [7]



Restructured silica (green) and soot[2] agglomerate d_m/d_p (red) as function of their $d_{m,o}/d_p$ measured by TDMA (filled symbols) and fitted to a scaling law (broken lines).



Mass-mobility relationships measured here by TDMA (lines) are validated against APM (symbols).

References

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[3] van Ommen J. (2012) *J. Nanopart. Res.* **14**, 737.
[4] Lee SY, Chang H, Ogi T, Iskandar F, Okuyama K. (2011) *Carbon* **49**, 2163.
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[6] Zangmeister CD, Radney JG, Dockery LT, Young JT, Ma XF, You RA, Zachariah MR. (2014) *PNAS* **111**, 9037.
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Conclusions

1. At S = 0.9, the ρ_{eff}/ρ of ramified silica agglomerates decreases with d_m/d_p, following closely the agglomeration theory.[5]
2. At S = 1.5, silica PPs restructure into spherical agglomerates with 10 % smaller $\bar{d}_m$ and ρ_{eff}/ρ = 0.28 ± 0.02 invariant of d_m/d_p.
3. Silica agglomerates consist of larger aggregates compared to those of soot due to their flame synthesis conditions. This results in less compact agglomerates with smaller ρ_{eff}/ρ and k_m after water condensation and evaporation.
4. TDMA measurements can be coupled with the scaling law derived here to measure the restructured agglomerate mass-mobility relationship as an alternative to the tedious APM.