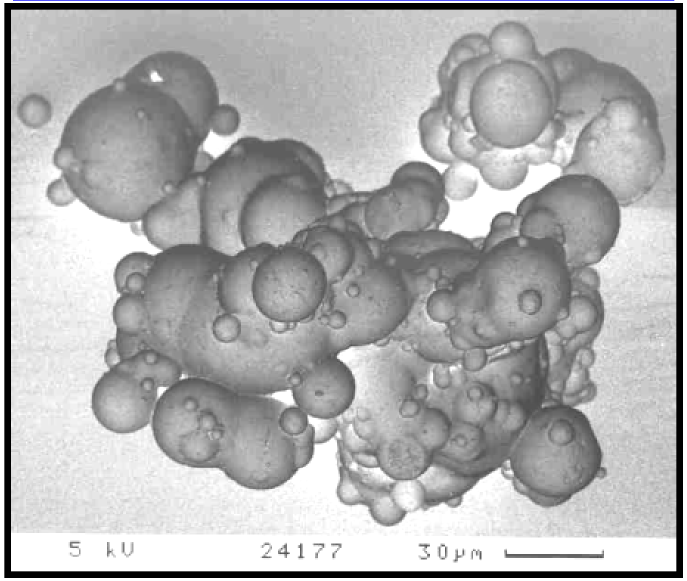
Lagrangian Agglomeration Models with Applications to Spray Drying

M. Sommerfeld and S. Stübing

Typical agglomerate from spray drying



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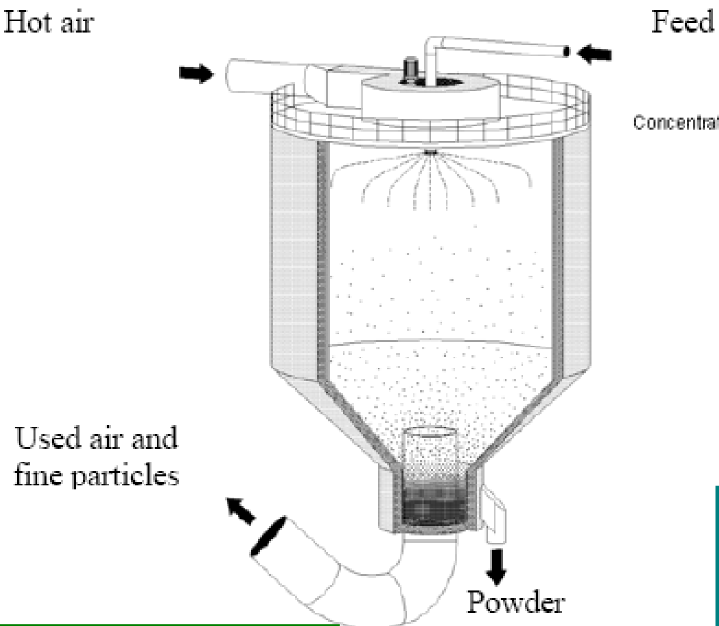
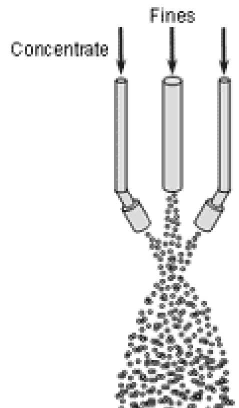
Content of the Lecture

- Summary of technical relevant agglomeration processes
- Introduction otoagglomerate properties
- Flow model; homogeneous isotropic turbulence
- Lagrangian particle tracking and turbulence dispersion model
- Lagrangian stochastic collision model
- Agglomeration simple and structure model
- Test case simulations for dry agglomeration
- Agglomeration of viscous particles with penetration
- Preliminary spray dryer calculations
- Conclusions and outlook

Introduction

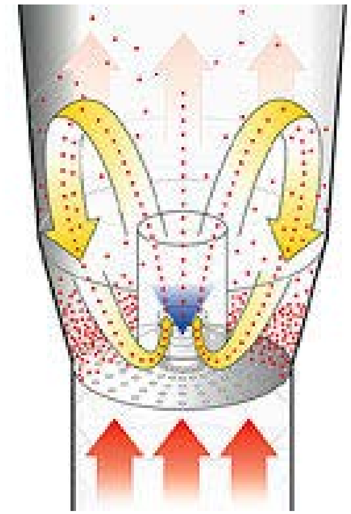
☞ **The agglomeration of particles is important for a number of technical and industrial processes in particle technology.**



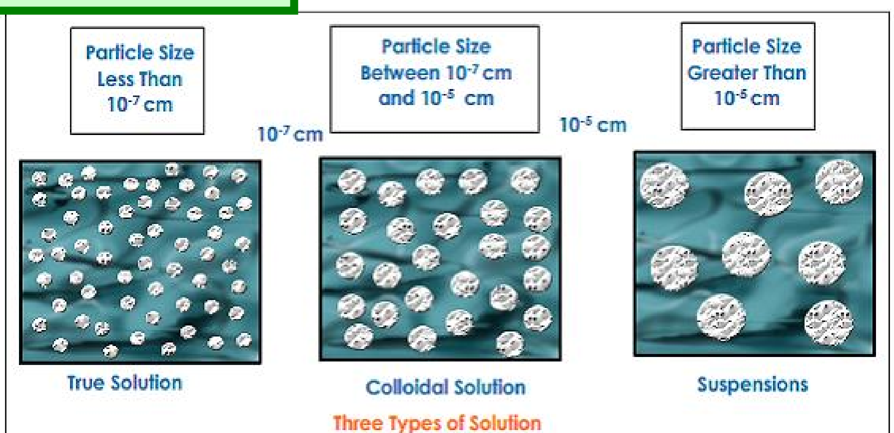


Spray drying and agglomeration

Fluidised bed granulation
Fluidised bed coating



Colloidal systems



Production of nano-particles by flame synthesis

Numerical Calculation of Particle-Laden Systems

☞ **For the numerical calculation of gas-solid flow systems the Euler/Lagrange approach is most favourable.**

Consideration of particle size distribution

Detailed modelling of the elementary processes

- **Particle-wall collisions**
- **Inter-particle collisions**
- **Agglomeration and breakage**

➢ **Sequential calculation of fluid flow and particle phase by a coupled hybrid approach until overall convergence is reached:**

- **Continuum fluid flow: Eulerian, RANS or LES approach with two-way coupling (droplet phase source terms)**
- **Particle phase simulated by the Lagrangian approach where a large number of representative particles (point-mass) are tracked through the flow field**

◆ Relevant forces on particles

◆ Modelling elementary processes

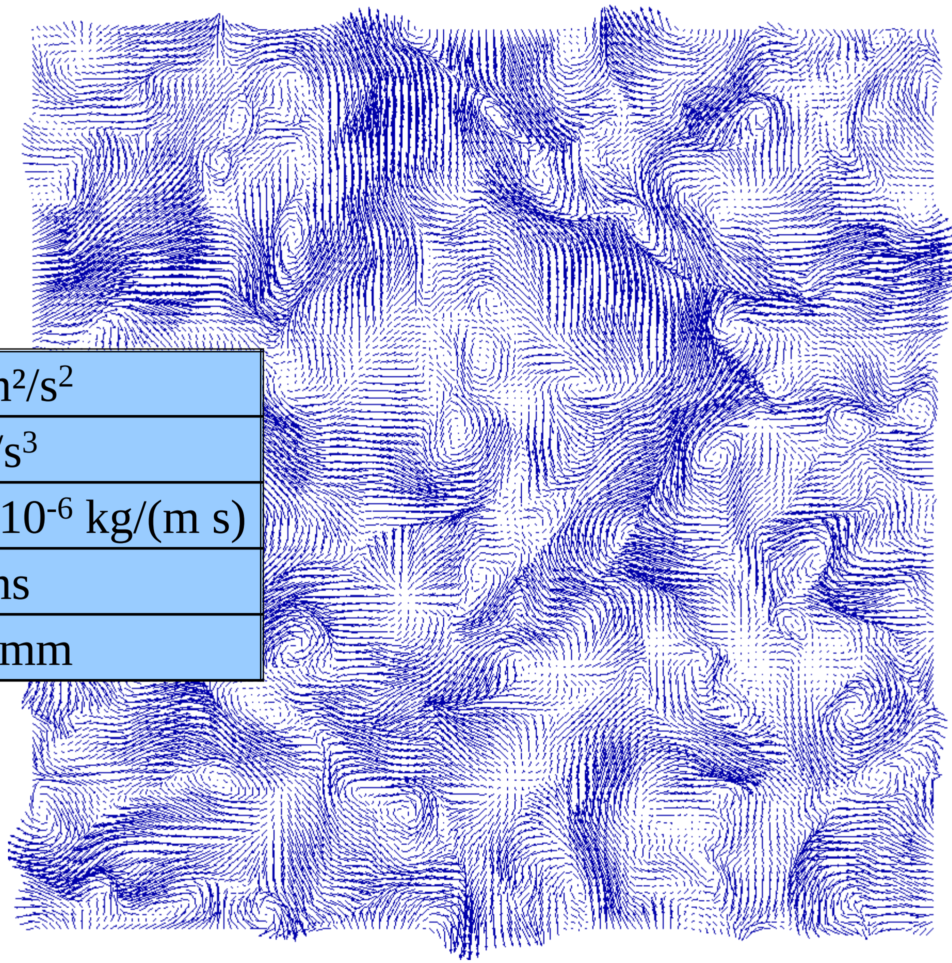
- ❖ Turbulent dispersion
- ❖ Particle collisions/agglomeration

Flow Conditions

- **The performance of the agglomerate structure model is tested for a pre-defined forced homogeneous isotropic turbulence (HIT) in a box**
- **Summary of turbulence properties**

LBM of HIT

$$T_L = 0.16\,\frac{k}{\varepsilon}; \quad L_E = 3.0 \cdot T_L$$



Turbulent kinetic energy k	0.06 m^2/s^2
Turbulent dissipation rate ε	10 m^2/s^3
Dynamic viscosity μ	18.23 10^{-6} kg/(m s)
Turbulent integral time scale T_L	0.96 ms
Lagrangian length scale L_E	0.576 mm

Lagrangian Particle Tracking

- **The Lagrangian approach requires the tracking of a large number of computational point-particles (parcels) solving the following equations:**

Particle position

$$\frac{d\vec{x}_p}{dt} = \vec{u}_p$$

Particle velocity (drag only)

$$m_p \frac{d\vec{u}_p}{dt} = \frac{\rho}{2} A_P C_D \left(\vec{u} - \vec{u}_P\right)\left|\vec{u} - \vec{u}_P\right|$$

$$C_D = \frac{24}{Re_p}\left(1 + 0.15\, Re_p^{0.687}\right) \quad \text{with } Re_p = \frac{\rho D_p \left|\overline{u} - \overline{u}_p\right|}{\mu}$$

- **The drag coefficient and the drag force of agglomerates is calculated for a sphere with diameter of Gyration.**
- **The porosity of the agglomerate is not accounted for in the drag coefficient.**
- **The instantaneous fluid velocity is generated by a single-step Langevin model.**

$$\boxed{u^f_{i,n+1} = R_{P,i}(\Delta t, \Delta r)\, u^f_{i,n} + \sigma_i \sqrt{1 - R^2_{P,i}(\Delta t, \Delta r)}\, \xi_i}$$

- **Particle rotation is not accounted for explicitly.**
- **10,000 primary particles are initially randomly distributed in the computational domain.**

Collision and Agglomeration Modelling

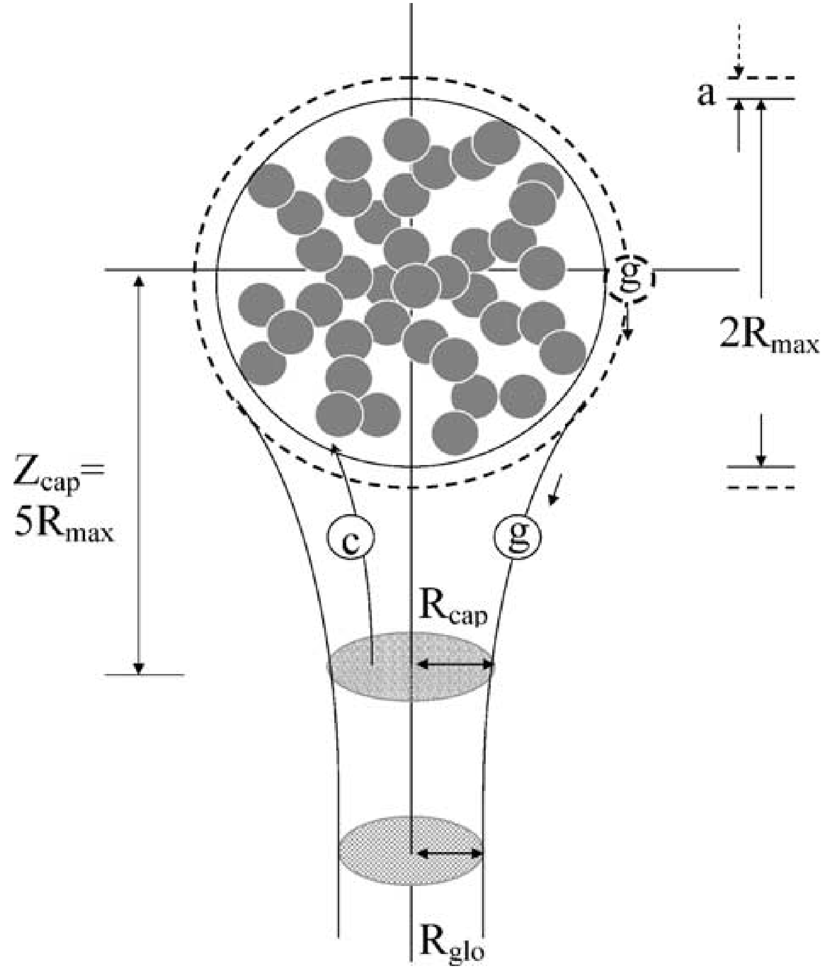
- In modelling agglomeration processes (**collision of primary particles with agglomerates**) several physical phenomena have to be accounted for:

❶ Detection of a possible collision with the stochastic collision model by defining the collision cross-section of an agglomerate

❷ Consideration of the impact efficiency (the primary particle might move with the relative flow around the agglomerate)

❸ The primary particle might fly through the dendrite branches of the agglomerate (hit probability)

❹ Which primary particle in the agglomerate is the collision partner of the new primary particle ???



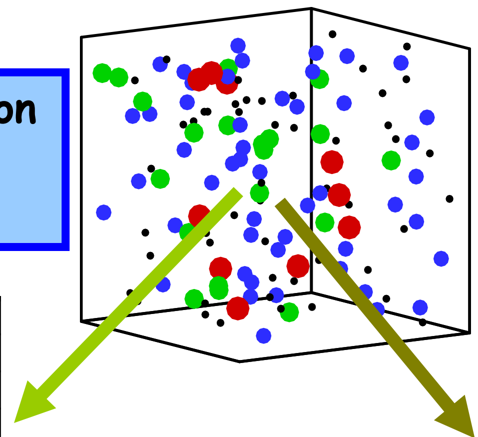
❺ Are the two primary particle sticking together considering adhesion forces or penetration ???

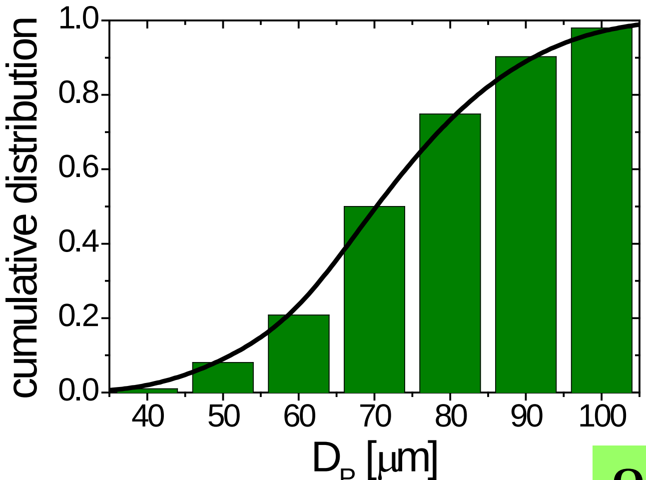
Stochastic Inter-Particle Collision Model 1

➲ Stochastic Inter-Particle Collision Model (Sommerfeld 2001)

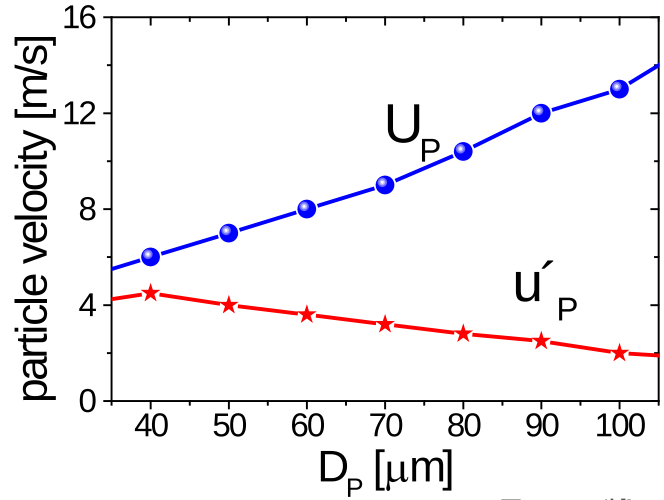
- **In the trajectory calculation of the considered particle (parcel) a fictitious collision partner is generated for each time step.**
- **The properties of the fictitious particle (representative of local population) are sampled from local, cell-based distribution functions:**

⇨ Particle number concentration
⇨ Particle diameter
(locally sampled distribution)





⇨ Particle velocities
(size-velocity correlations)
sampled from normal PDF



Q_n > RN [0, 1]

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Stochastic Inter-Particle Collision Model 2

- In turbulent flows the sampled fictitious particle velocity fluctuation is correlated with that of the real particle:

$$u'_{fict,i} = R(\tau_P, T_L)\, u'_{real,i} + \sigma_i \sqrt{1 - R(\tau_P, T_L)^2}\; \xi_n$$

$$R(\tau_p, T_L) = \exp\left(-0.55 \left(\frac{\tau_p}{T_L} \right)^{0.4} \right)$$

Resulted from comparison with LES (group of Prof. Simonin)

- Calculation of collision probability between the considered particle and the fictitious particle:

$$P = \frac{\pi}{4} (D_{P1} + D_{P2})^2 \left| \vec{u}_{P1} - \vec{u}_{P2} \right| n_P \; \Delta t$$

- A collision occurs when a random number in the range [0 - 1] becomes smaller than the collision probability.
- The collision process is calculated in a co-ordinate system where the fictitious particle is stationary (central oblique collision).
- Generation of impact point on an equivalent sphere of the agglomerate by a random process and determination of the impact parameter L:

$$L = \sqrt{Y^2 + Z^2} \quad \text{with:} \quad L \le 1$$
$$\phi = \arcsin(L)$$

$$0 < \Psi < 2\,\pi$$

Stochastic Inter-Particle Collision Model 3

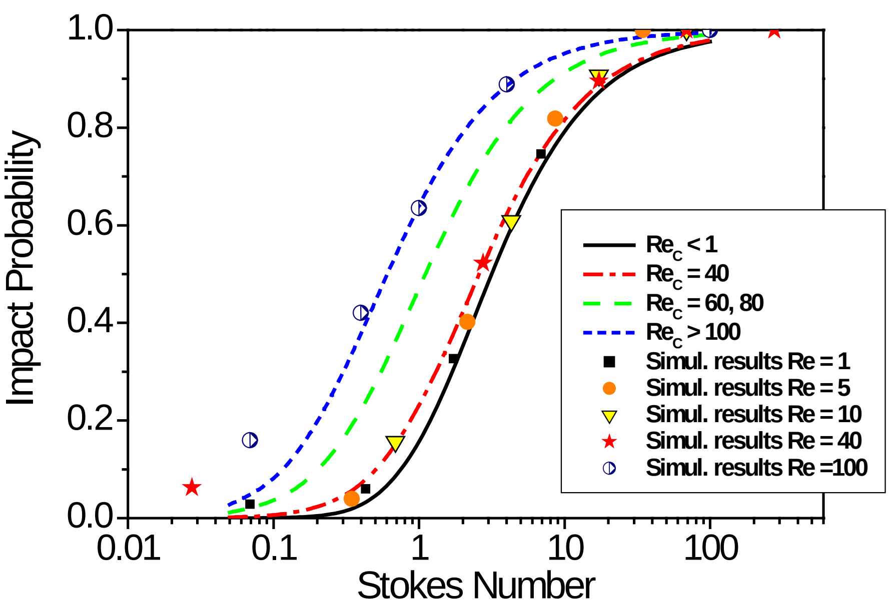
- **Consideration of fluid dynamic effects for the interaction of particles with different size (impact efficiency), see Ho and Sommerfeld (2002).**

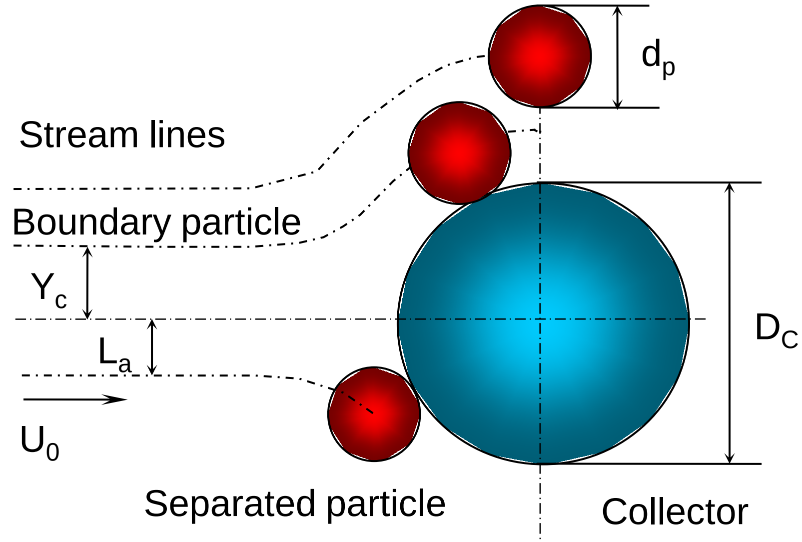
For the inertial regime the impact efficiency may be calculated from (Schuch and Löffler 1978):

$$\eta = \left(\frac{2\, Y_C}{D_C} \right)^2 = \left(\frac{St_i}{St_i + a} \right)^b$$

$$St_i = \frac{\rho_p\, d_p^2 \left| \vec{u}_p - \vec{u}_C \right|}{18\, \mu\, D_C}$$

Re_C	a	b
>> 1	0.25	2.0
60 – 80	0.506	1.84
40	1.03	2.07
10 – 20	1.24	1.95
< 1	0.65	3.7





collision if: $L_a \le Y_C + \dfrac{d_p}{2}$

Stochastic Inter-Particle Collision Model 4

- **In the case of rebound the new velocities of the considered particle are calculated by solving the momentum equations for an oblique collision in connection with Coulombs law of friction.**

$$u'_{P1} = u_{P1}\left(1 - \frac{1+e}{1+m_{P1}/m_{P2}}\right)$$

Non-sliding collision

$$\frac{v_{P1}}{u_{P1}} < \frac{7}{2}\mu\left(1-e\right)$$

Sliding collision

$$v'_{P1} = v_{P1}\left(1 - \mu\left(1+e\right)\frac{u_{P1}}{v_{P1}}\frac{1}{1+m_{P1}/m_{P2}}\right)$$

$$v'_{P1} = v_{P1}\left(1 - \frac{7/2}{1+m_{P1}/m_{P2}}\right)$$

- **Re-transformation of the new particle velocities in the laboratory frame of reference.**
- **Particle rotation is not considered in agglomeration studies, due to the complex momentum exchange for structured agglomerates.**

Agglomeration Model for Solid Particles 1

☞ **The occurrence of agglomeration may be decided on the basis of an energy balance (dry particles → only Van der Waals forces):**

$$E_{k1} \leq \Delta E_{vdw} + E_d$$

$$E_d = \left(1 - k_{pl}^2\right) E_{k1}$$

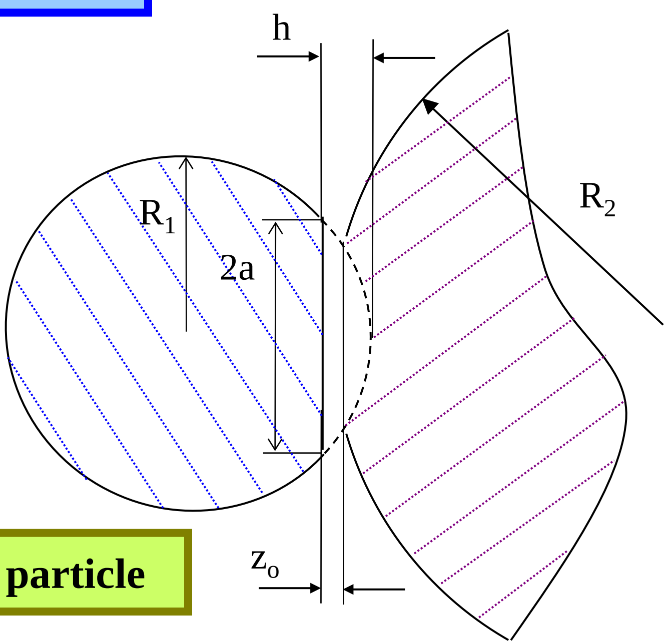
Ho and Sommerfeld (2002).

Van der Waals Energie:

$$\Delta E_{vdw} = -\int_{z_0}^{\infty} \frac{A}{6\,\pi\, z^3}\, \pi\, a^2\, dz$$

Restitution ratio:

$$k_{pl}^2 = \frac{E_{k1} - E_d}{E_{k1}}$$



R1: smaller particle

☞ **Critical impact velocity:**

$$\left|\vec{U}_{kr}\right| = \frac{1}{2\,R_1}\, \frac{\left(1 - k_{pl}^2\right)^{1/2}}{k_{pl}^2}\, \frac{A}{\pi\, z_0^2\, \sqrt{6\, P_{pl}\, \rho_p}}$$

Agglomeration if:

$$\vec{U}_{rel}\, \cos\phi \leq \left|\vec{U}_{kr}\right|$$

Agglomeration Model for Solid Particles 2

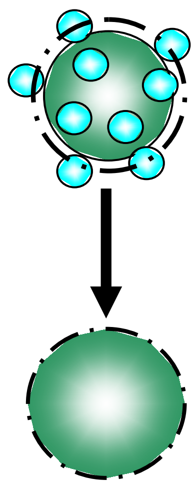
Sommerfeld & Stübing
ETMM 9, 2012

Agglomeration models

Point-particle assumption

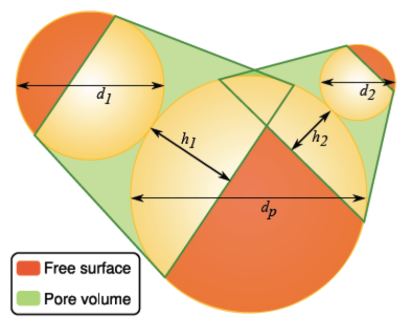
Simple agglomeration model

Volume equivalent sphere



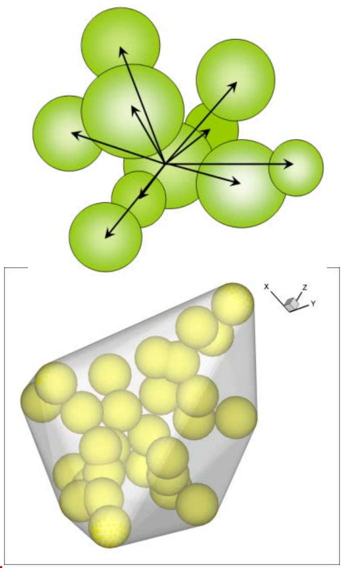
- Number of primary particles
- Penetration depth

Sequential agglomeration model



- Number of primary particles
- Hull volume/diameter
- Porosity of hull
- Contact forces

Agglomerate structure model



Location vectors
Convex hull

$$\varepsilon = 1 - \frac{V_{Part}}{V_{Hull}}$$

- Agglomerate structure
- Effective surface area
- Volume of convex hull
- Porosity of the agglomerate

Agglomeration Model for Solid Particles 3

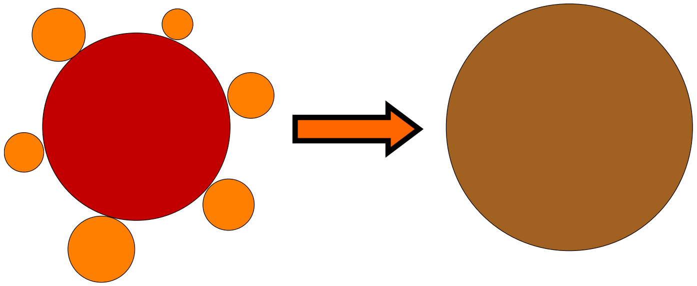
☞ **Through the comparison of the critical velocity with the normal impact velocity two types of collisions may occur:**

Rebound:

The solution of the impulse equations in connection with Coulombs law gives the rebound velocities of the considered particle.

Agglomeration:

A new particle is formed with the volume equivalent diameter.



$$d_{agglo} = \sqrt[3]{d_{real}^3 + d_{fict}^3}$$

The number of real particles is updated in order maintain the total mass in the parcel.

$$n_{agglo} = n_{real} \frac{d_{real}^3}{d_{real}^3 + d_{fict}^3}$$

The new velocity of the agglomerate follows from a momentum balance.

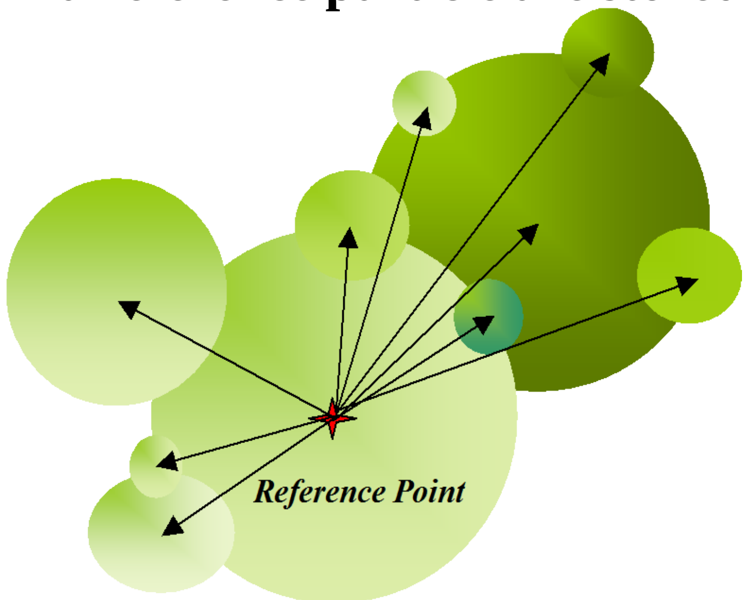
$$u_{agglo,i} = u_{real,i} \frac{m_{real}}{m_{real} + m_{fict}} \quad i = 1, 2$$

➲ **In the subsequent particle tracking forces are calculated based on the volume equivalent sphere (simplification, no other properties available).**

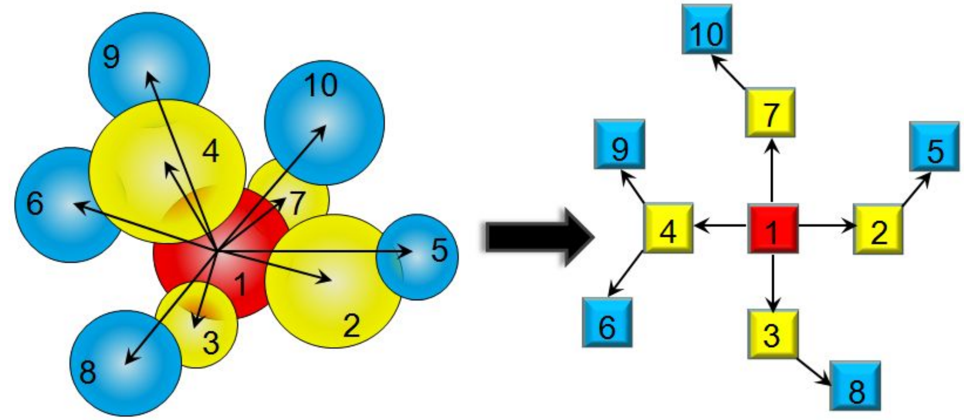
➲ **The collision probability is also calculated with the volume equivalent sphere.**

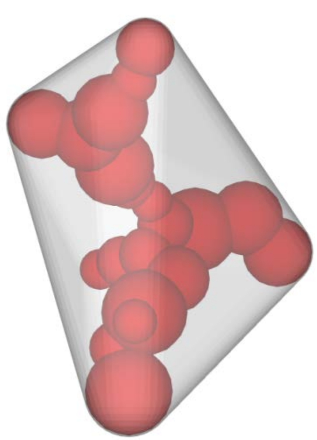
Agglomerate Structure Model 1

☞ In order to obtain more detailed information on the agglomerate structure, location vectors for all primary particles in the agglomerate with respect to a reference particle are stored.



The agglomerate structure is stored in a linked list





The agglomerate is still treated as point-particle !!!

Most important agglomerate properties

- Porosity of the agglomerate (convex hull)
- Effective surface area
- Agglomerate structure; Shape indicators

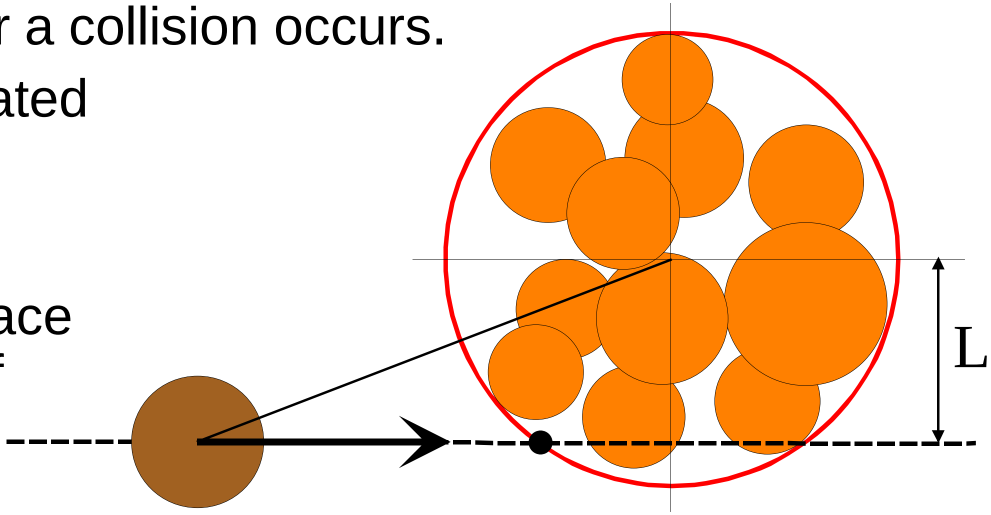
Agglomerate Structure Model 2

☞ **Assumptions for the stochastic collision model with respect to structure modelling:**

- Agglomerates can only collide with primary particles (number concentration of the resulting agglomerates is very low).
- The fictitious particle cannot be an agglomerate, hence it is only sampled from the primary particle size distribution.

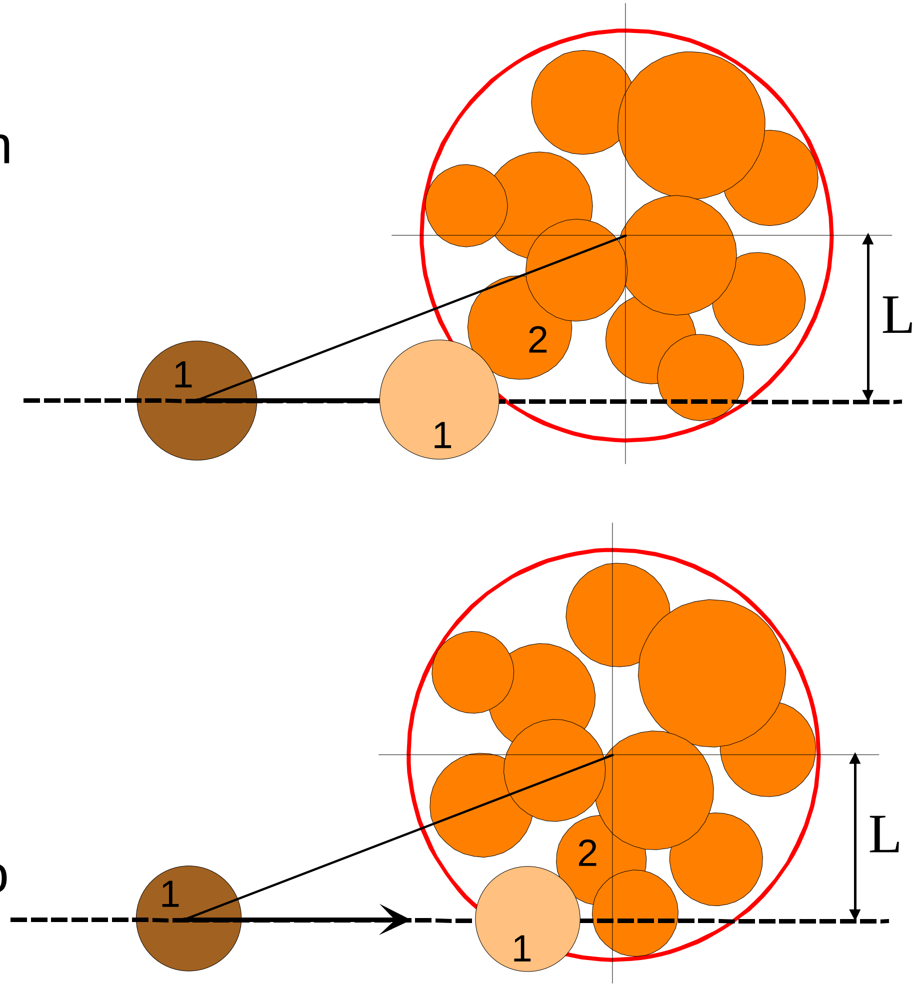
☞ **Extension of the stochastic collision model:**

- The collision probability (based on a selected collision sphere of the agglomerate) predicts whether a collision occurs.
- The collision process is calculated in a coordinate system where the agglomerate is stationary.
- The point of impact on the surface of the selected collision sphere of the agglomerate is sampled stochastically.



Agglomerate Structure Model 3

- A collision occurs if the lateral displacement *L* is smaller than the boundary trajectory Y_C (impact efficiency).
- Random rotation of the agglomerate in all three directions (since rotation is neglected).
- The particle then collides with the primary particle in the agglomerate being closest to the impact point (tracking).
- The critical velocity for the two involved primary particles is calculated.



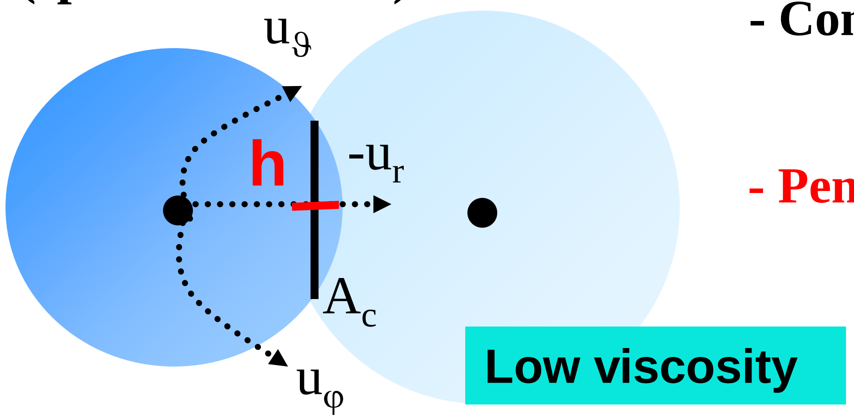
sticking

rebound

Viscous particles: penetration

Penetration Model for High Viscous Droplets

High viscous droplets penetrates into the low viscous droplet (spherical frame)



- Contact Area: $$d_{cont} = 2 \cdot \sqrt{h \cdot d_{Low} - h^2}$$

- Penetration depth $$X_P = \frac{d_{High} + d_{Low}}{2} - r$$

$$h = X_P \quad \text{for} \quad r > 0$$

$$h = \frac{d_{Low}}{2} - r \quad \text{for} \quad r \leq 0$$

➢ Calculation of ***time-dependent penetration depth:***

Radial:

➢ Motion of sphere in viscous liquid

$$m_{High} \cdot \frac{du_r}{dt} = -3 \cdot \pi \cdot \mu_{Low} \cdot d_{cont} \cdot u_r$$

$$\frac{dr}{dt} = u_r$$

Tangential:

➢ Shear force across contact area

$$m_{High} \cdot \frac{du_\vartheta}{dt} = -\mu_{Low} \cdot d_{cont} \cdot u_\vartheta$$

$$\frac{d\vartheta}{dt} = \frac{u_\vartheta}{r}$$

Dry Particle Agglomeration

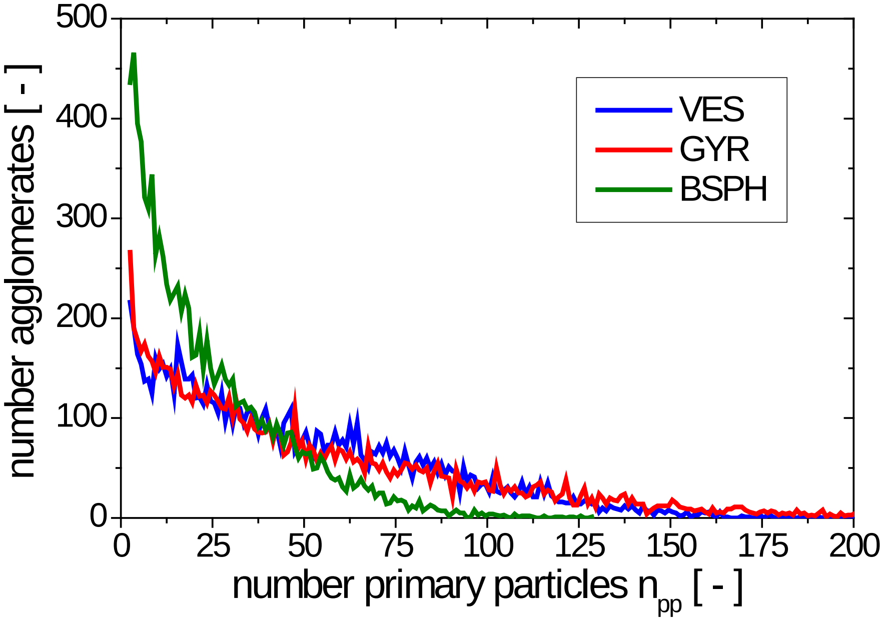
- Dry particle agglomeration (i.e. without penetration) is simulated for mono-sized particles and a size distribution with the following properties:

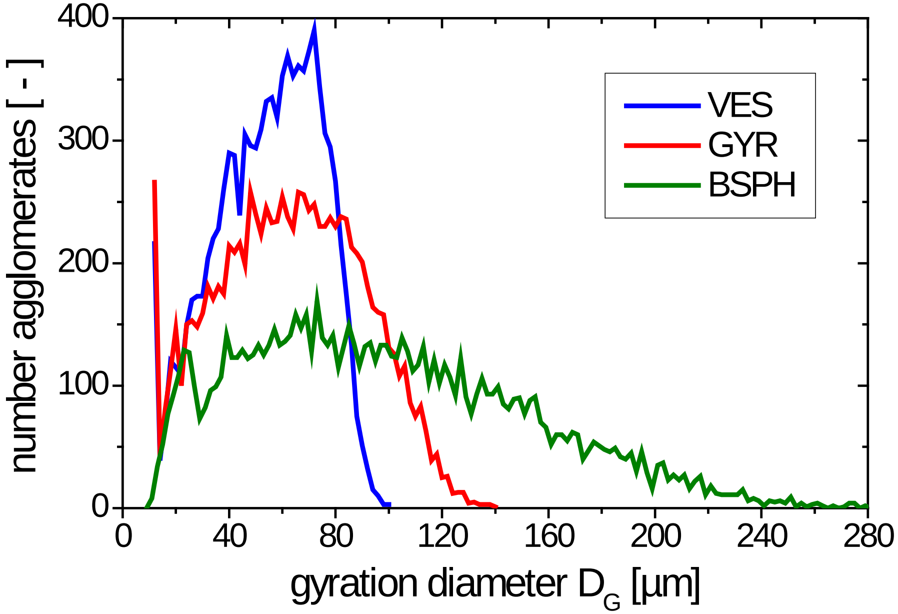
Primary particle diameter	5 µm	12 µm	2 – 20 µm
Particle density	1,000 kg/m³	1,000 kg/m³	1,000 kg/m³
Particle relaxation time τ_p	$0.076 \cdot 10^{-3}$ s	$0.432 \cdot 10^{-3}$ s	$(0.013 – 1.2) \cdot 10^{-3}$ s
Stokes-number $St = \tau_p / T_L$	0.08	0.45	0.013 – 1.3
Volume fraction	$1.0 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$

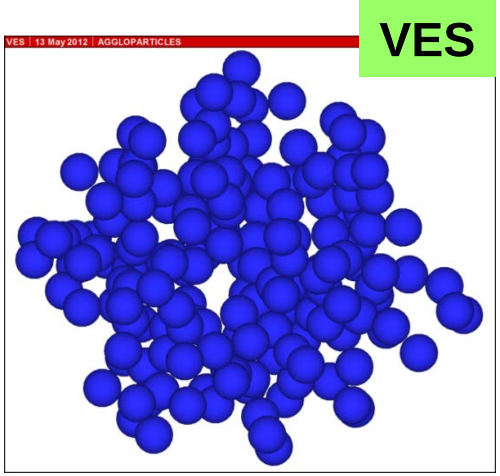
Hamaker Constant A	$5.0 \cdot 10^{-19}$ J
Restitution Ratio k_{pl}	0.6
Friction Coefficient µ	0.4
Limiting Pressure P_{pl}	$5.0 \cdot 10^{9}$ Pa
Minimum Contact Distance	40 nm

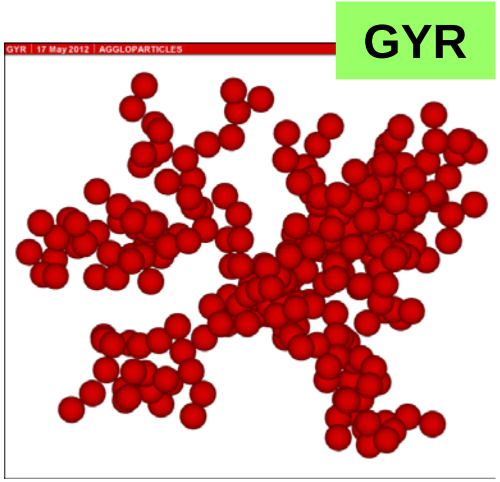
Dry Agglomeration, Mono 12 µm

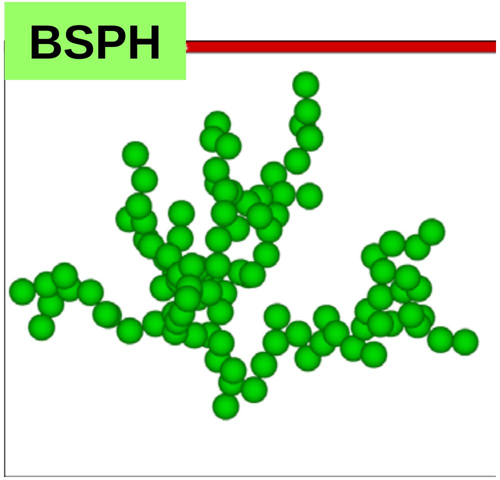
☞ **Agglomerate structure mono-sized 12 µm, without impact efficiency**





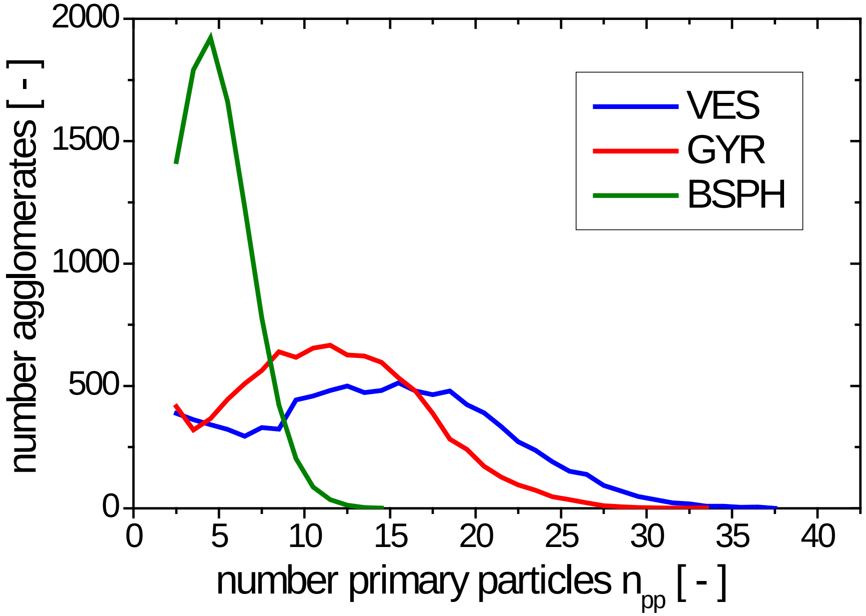


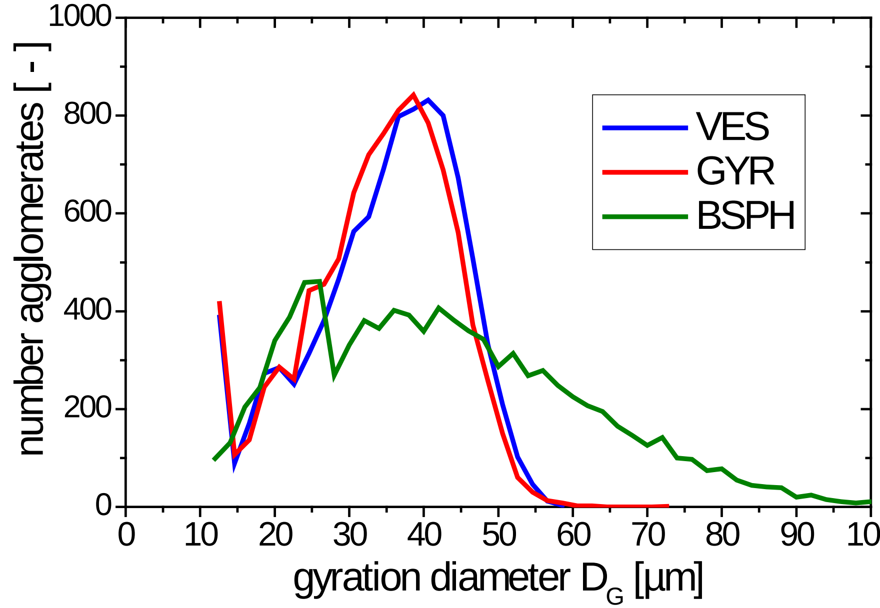


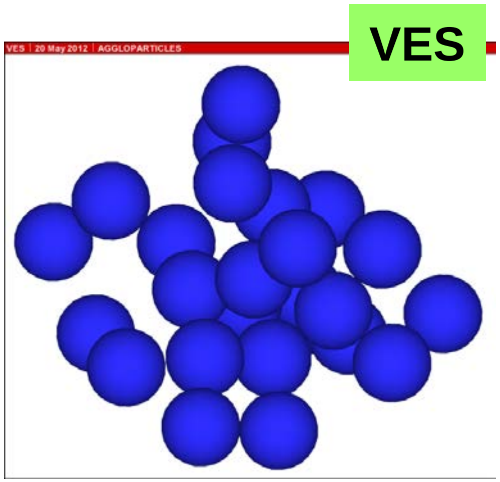


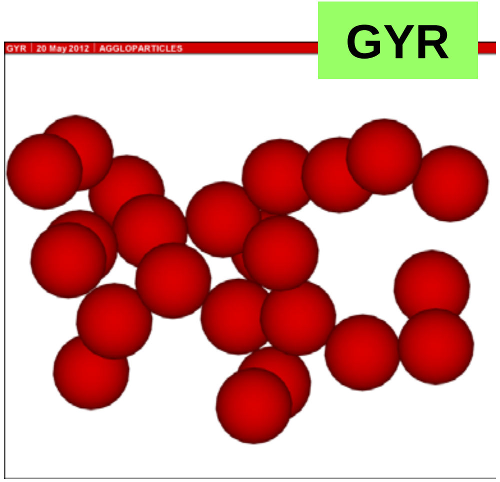
Dry Agglomeration, Mono 12 µm

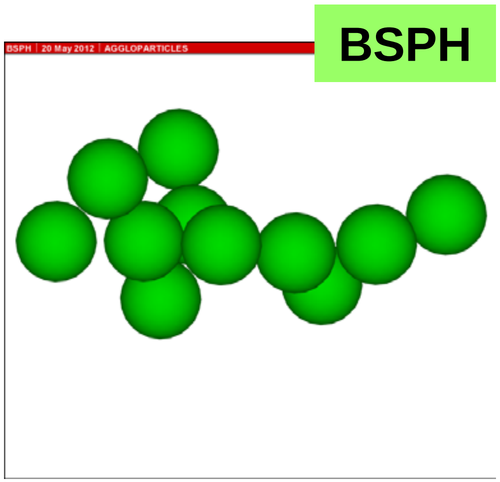
☞ **Agglomerate structure mono-sized 12 µm, with impact efficiency**





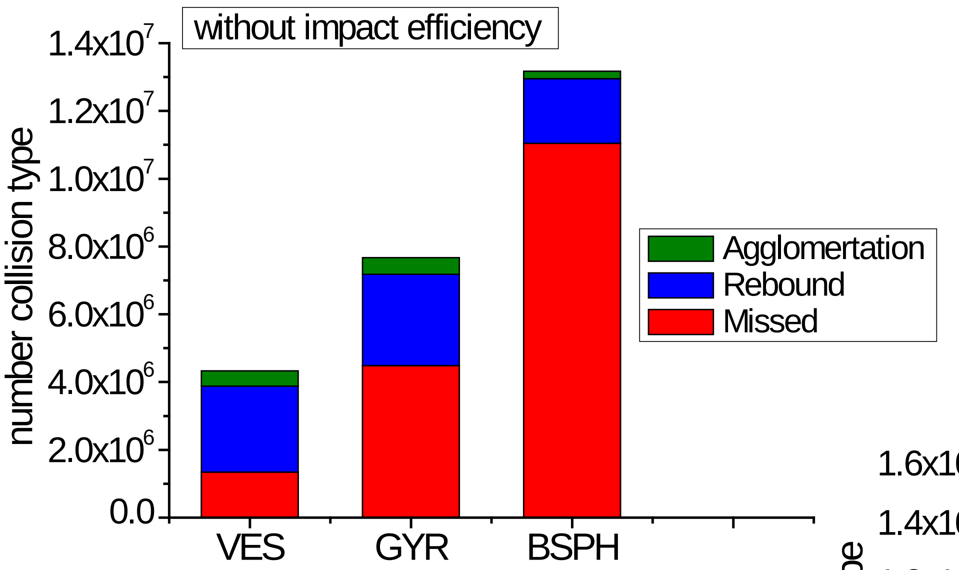


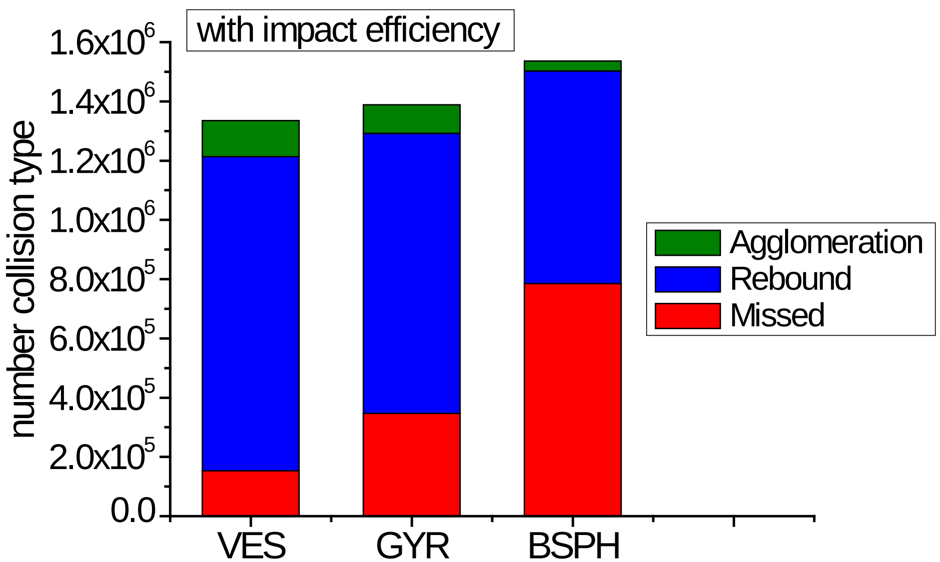




Dry Agglomeration, Mono 12 μm

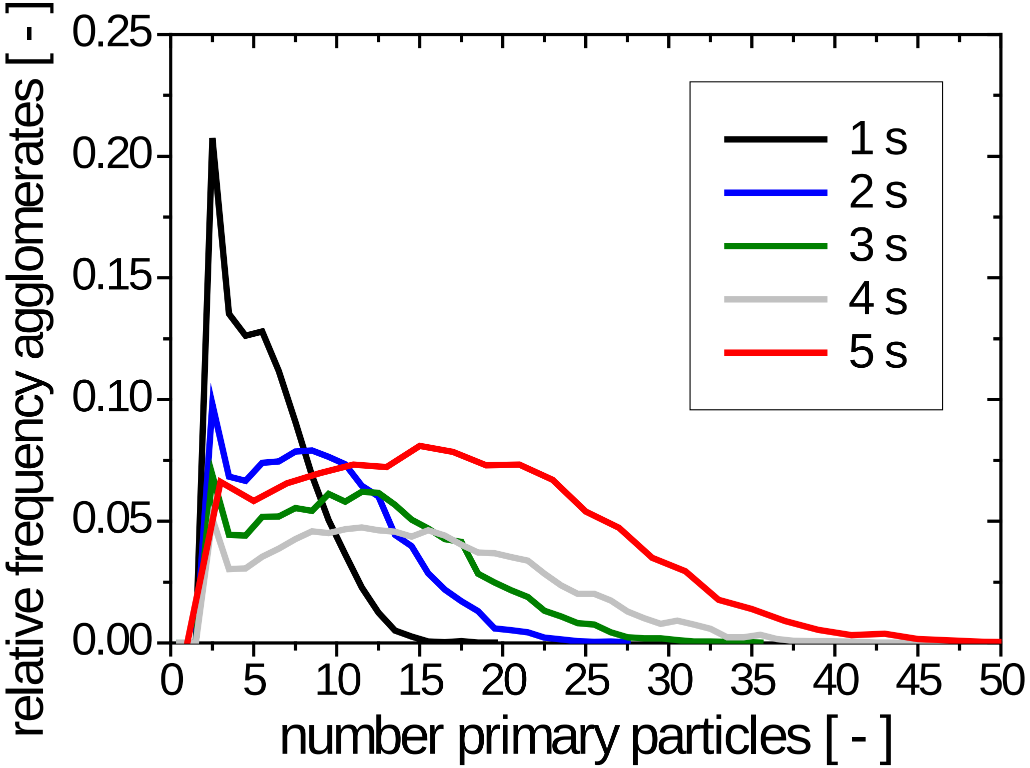
☞ **Number of collision types comparing without and with impact efficiency (mono-sized 12 μm)**

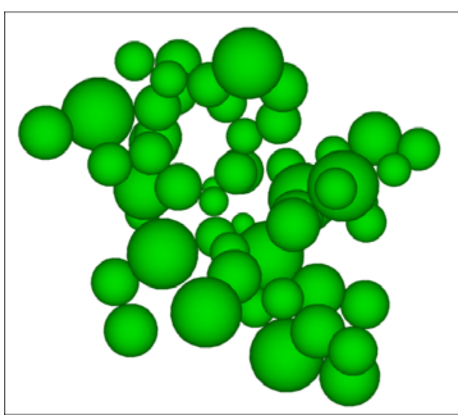


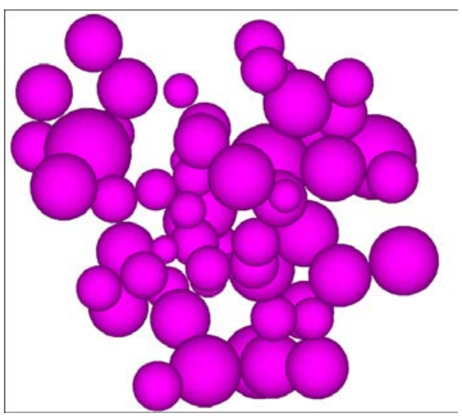


Dry Agglomeration, Distribution 2 – 20 µm

- **Temporal evolution of agglomerates for initial size distribution 2 – 20 µm.**

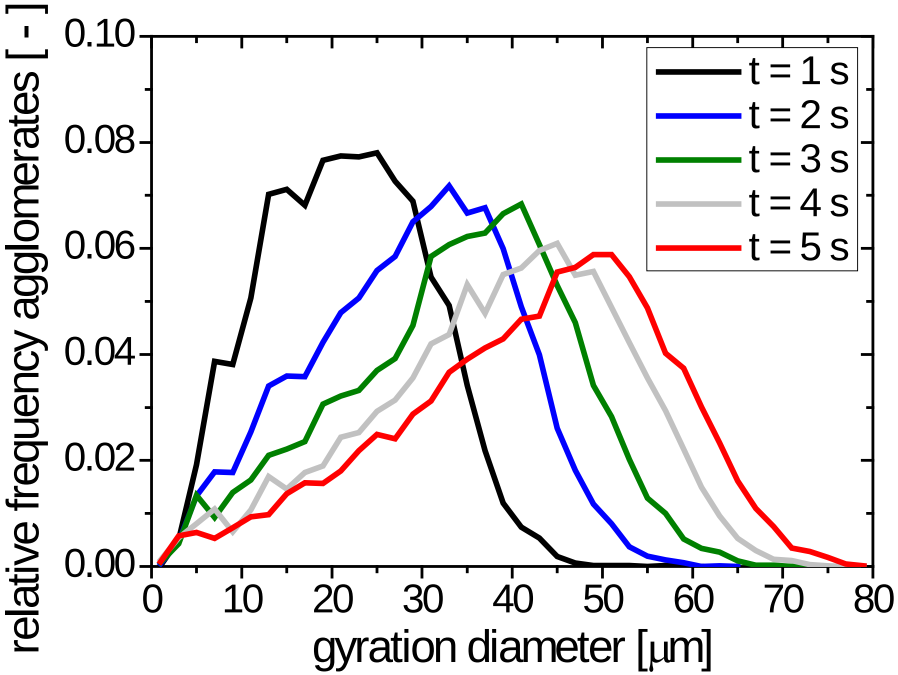


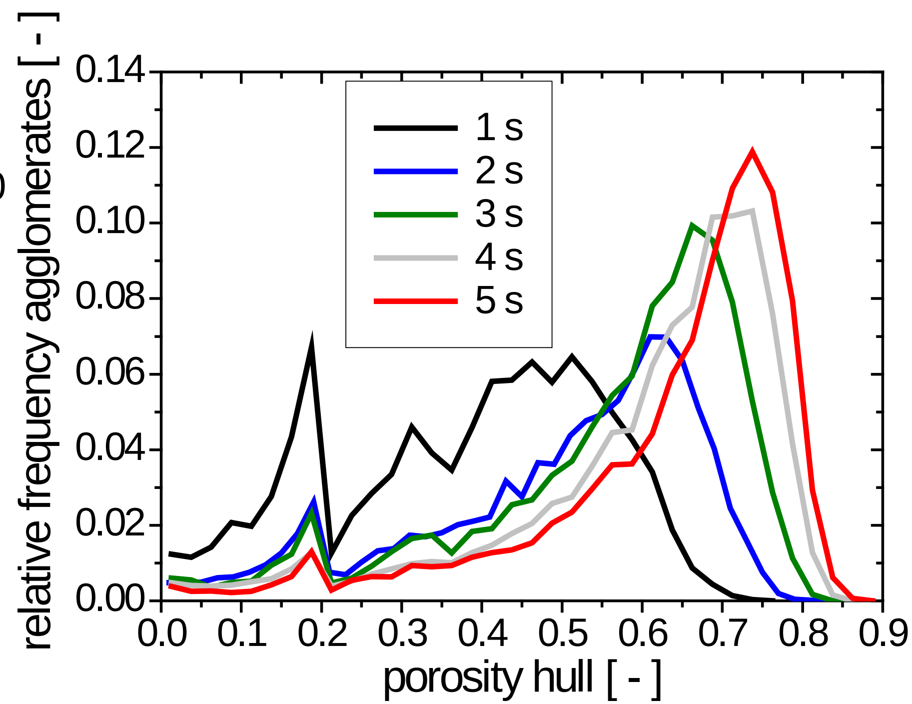




Dry Agglomeration, Distribution 2 – 20 μm

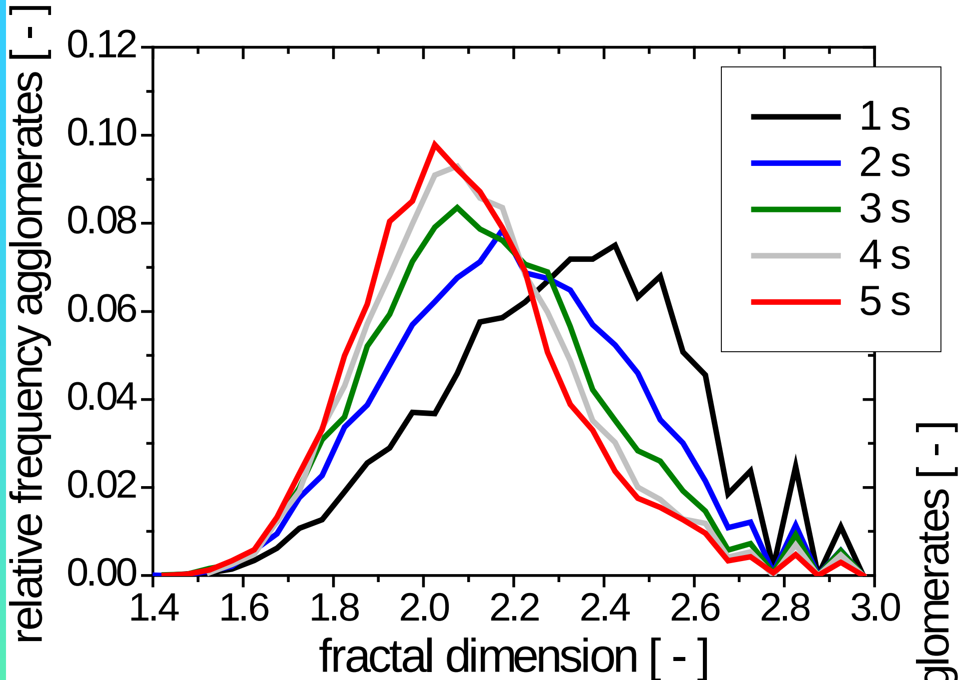
- Temporal evolution of agglomerates for initial size distribution 2 – 20 μm.

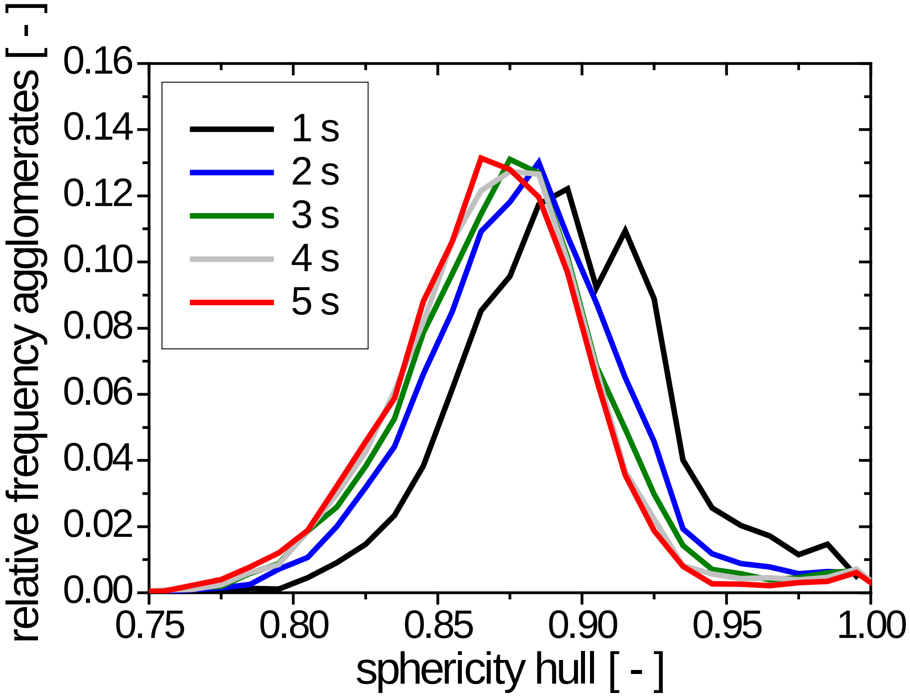




Dry Agglomeration, Distribution 2 – 20 μm

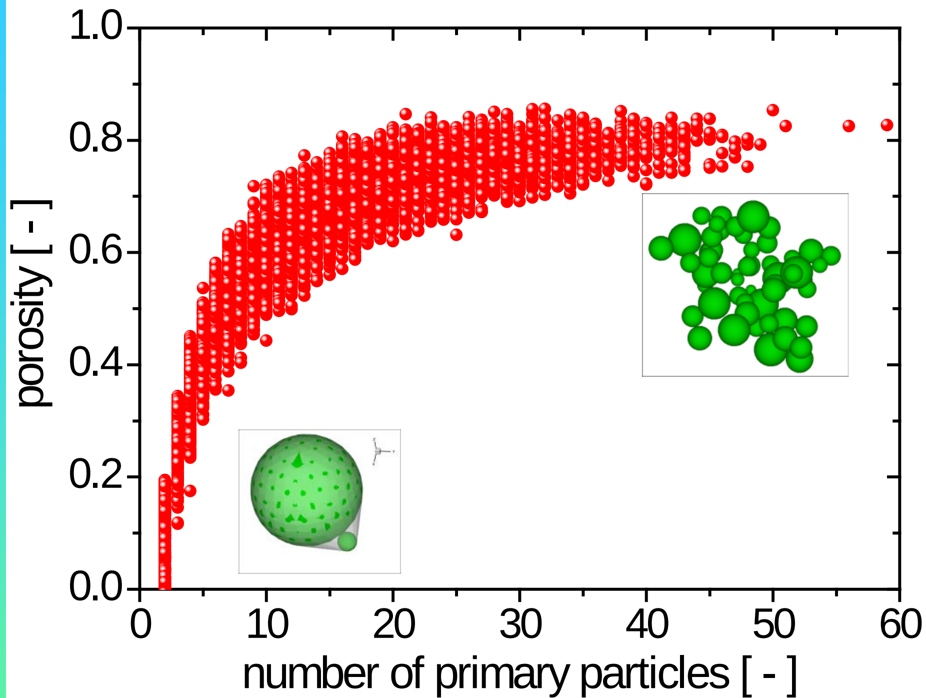
➢ **Temporal evolution of agglomerates for initial size distribution 2 – 20 μm.**

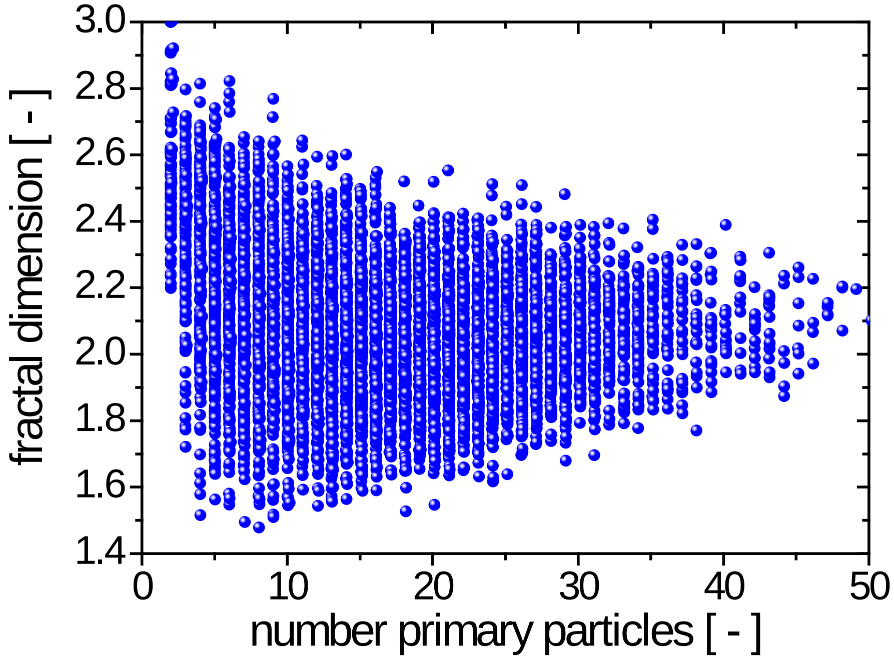




Dry Agglomeration, Distribution 2 – 20 μm

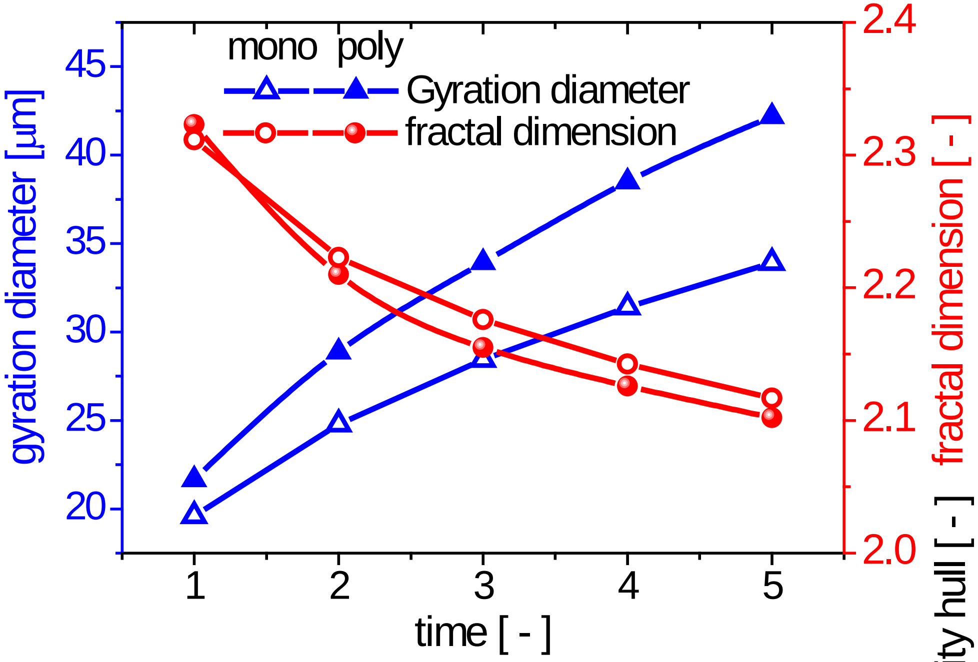
- **Properties in dependence of the number of primary particles in the agglomerate for initial size distribution 2 – 20 μm.**

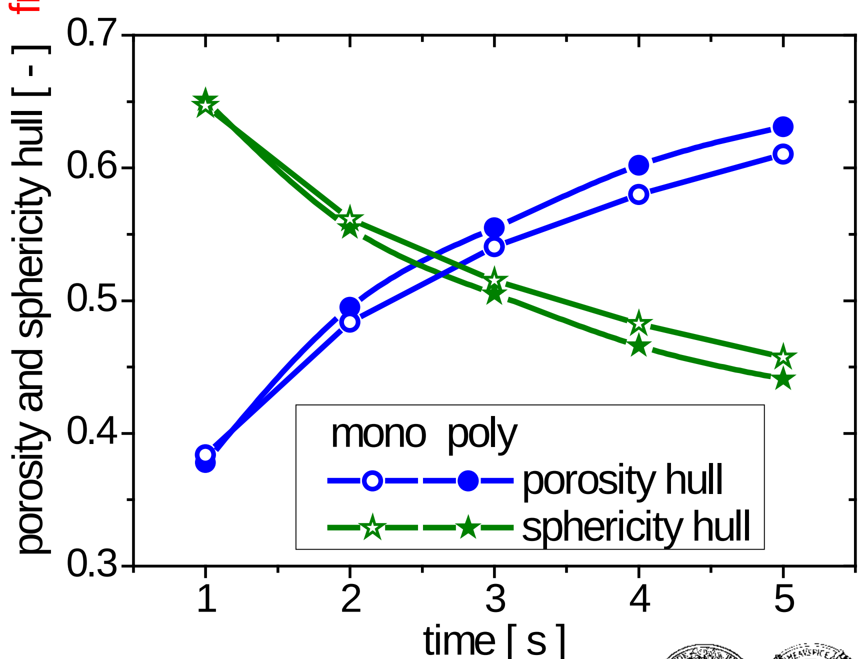




Dry Agglomeration, Distribution 2 – 20 µm

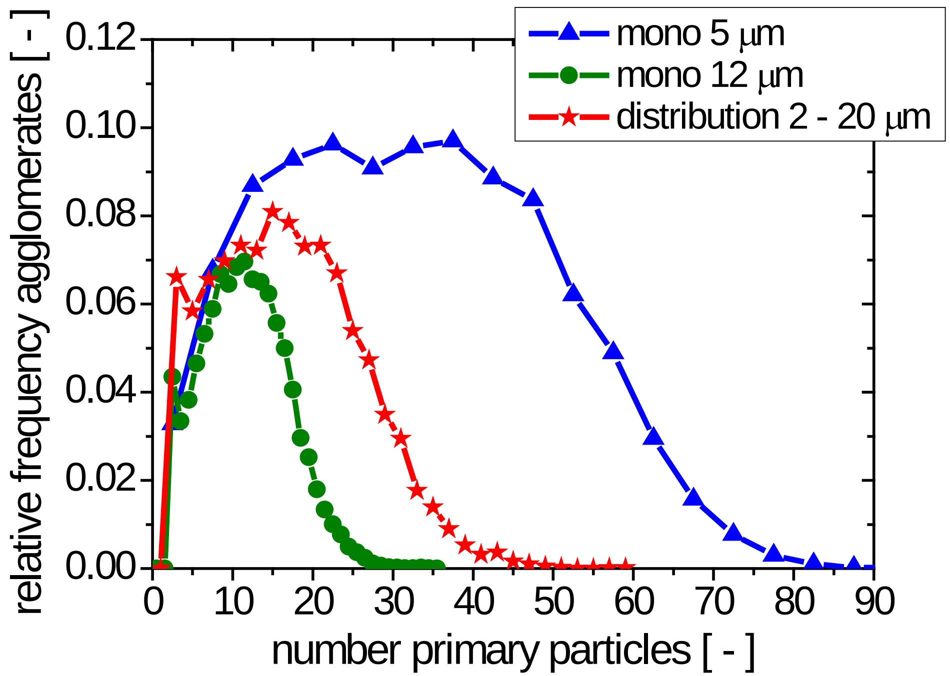
☞ **Temporal evolution of mean agglomerate properties, mono-sized 12 µm, poly-sized 2 – 20 µm**



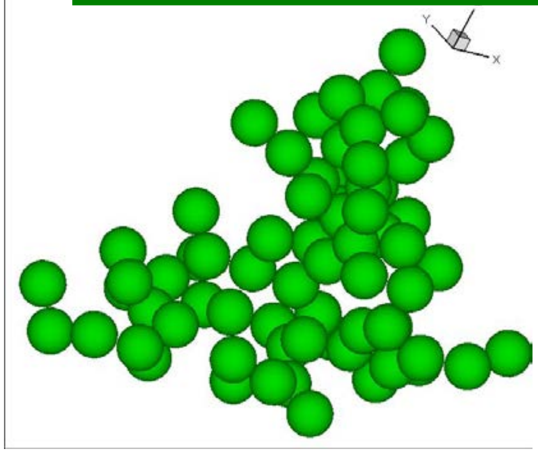


Comparison Initial Size

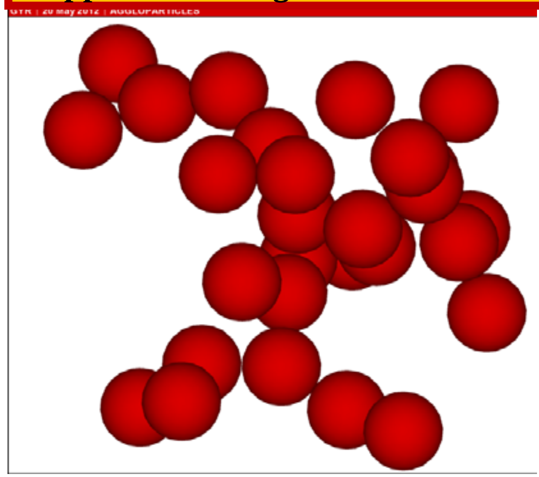
☞ **Comparison dry agglomeration all sizes after 5 seconds**



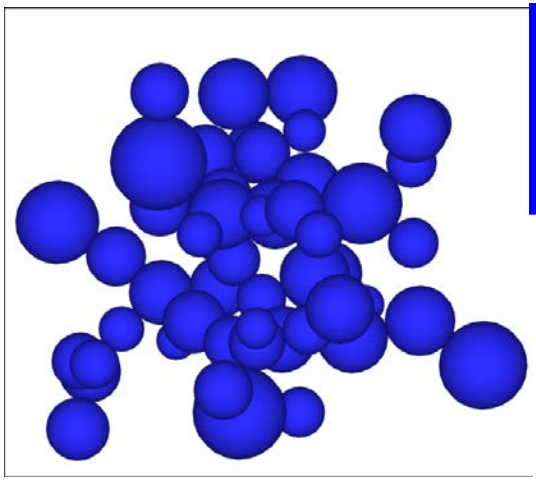
Mono-sized 5 μm
N_{pp} = 72; D_g = 31 μm; ε = 0.84



Mono-sized 12 μm
N_{pp} = 28; D_g = 52 μm; ε = 0.8

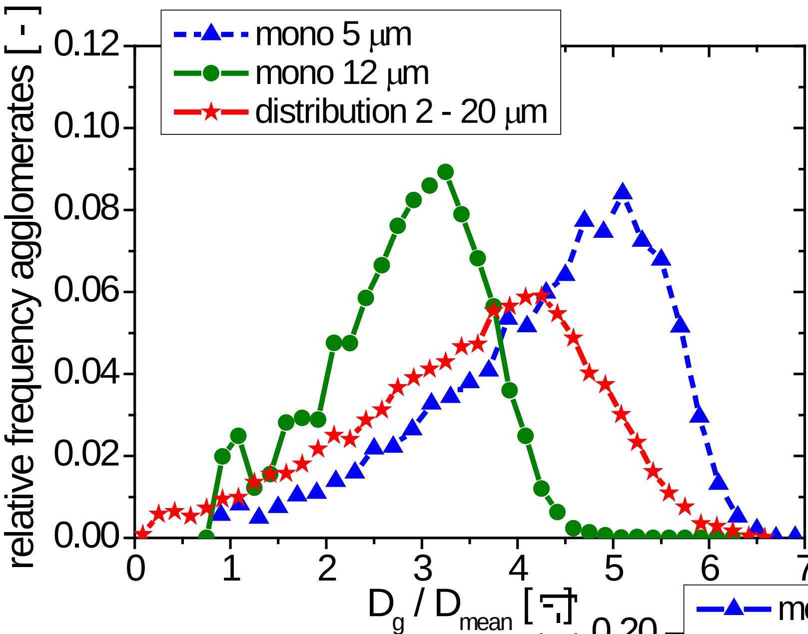


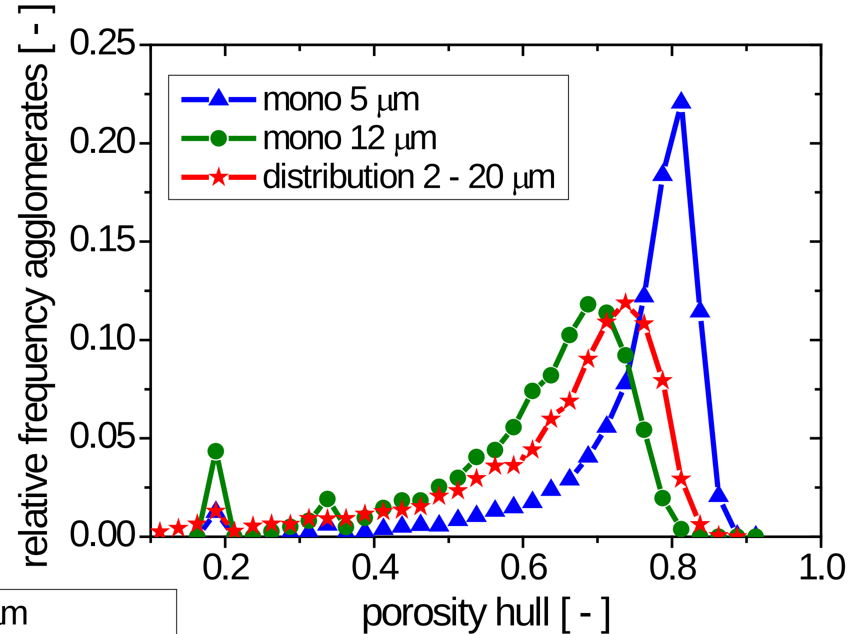
Distribution 2 – 20 μm
N_{pp} = 51; D_g = 68 μm; ε = 0.83

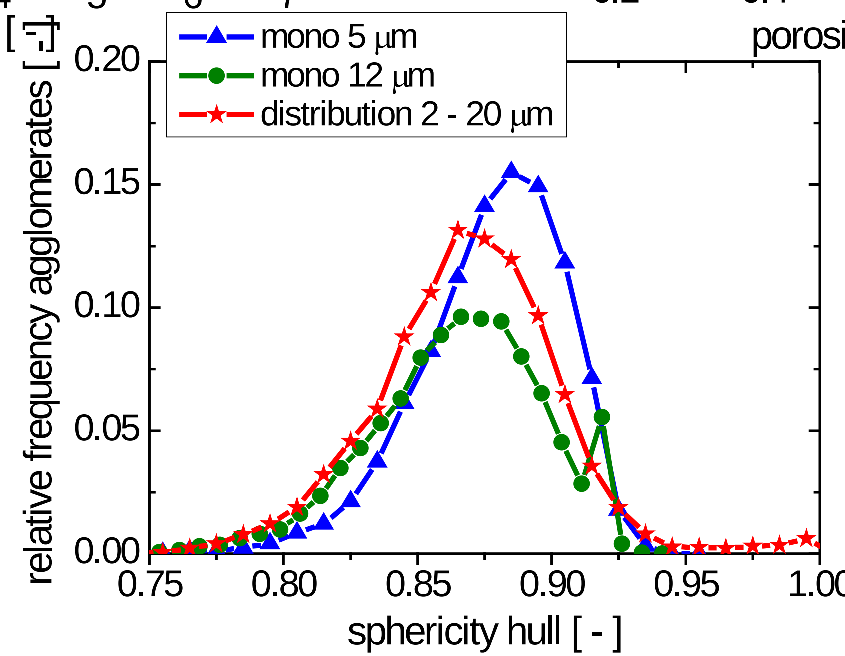


Comparison Initial Size

☞ Comparison dry agglomeration all sizes after 5 seconds

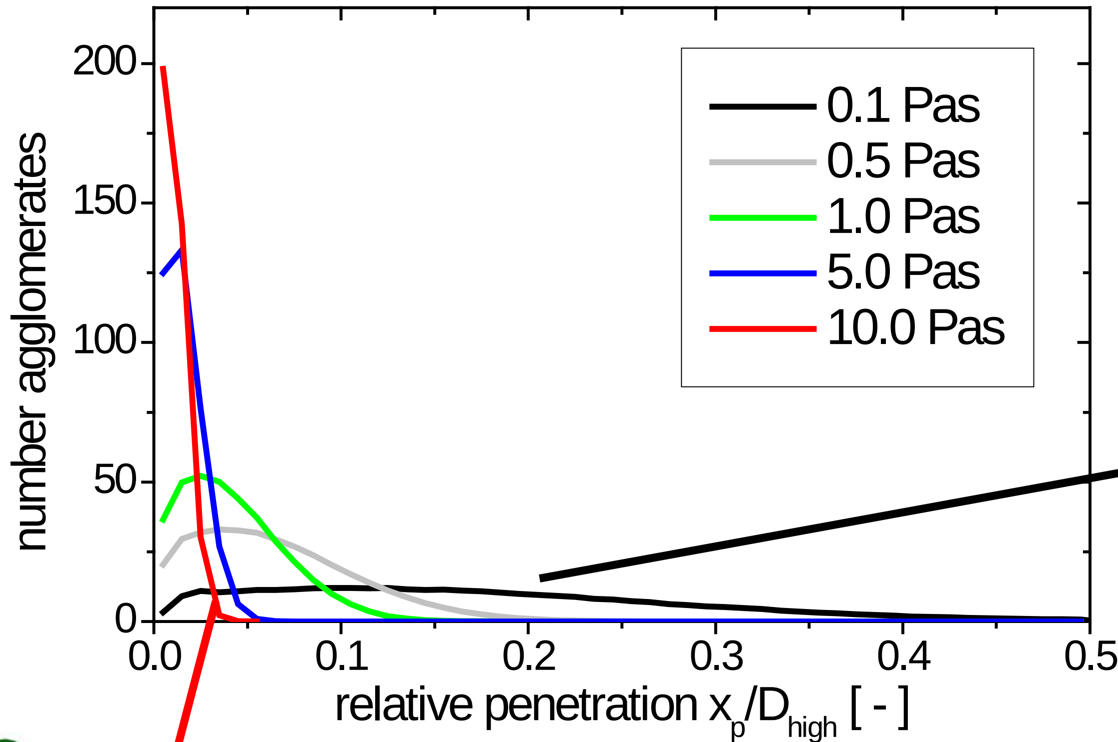


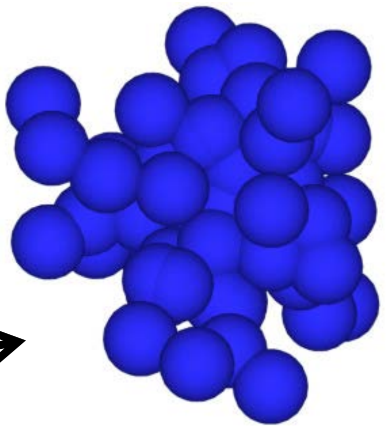


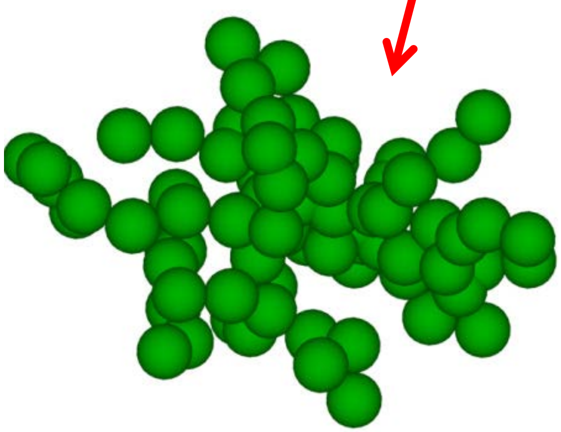


Viscous Particle Agglomeration

➲ **Properties of agglomerates with different viscosity mono-sized primary particles with 12 µm**

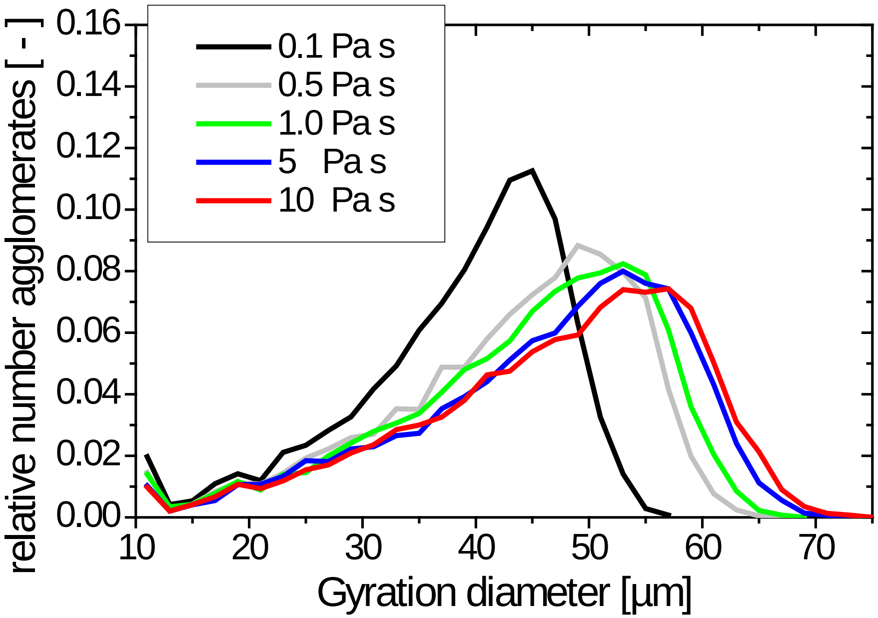


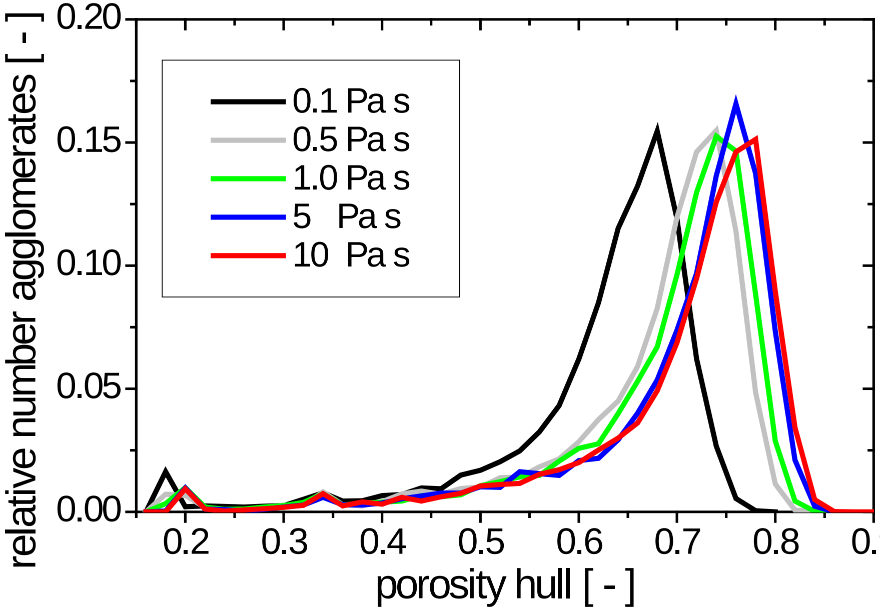




Viscous Particle Agglomeration

➲ **Properties of agglomerates with different viscosity, mono-sized primary particles with 12 µm**





Geometry of the Spray Dryer 1

➲ **Geometry and operational conditions of spray dryer (NIRO Copenhagen):**

Dryer geometry:

$H = 4096$ mm

$H_{cyl} = 1960$ mm

$D = 2700$ mm

$H_{out} = 3303$ mm

$D_{out} = 210$ mm

Annular Air Inlet

$R_o = 527$ mm

$R_i = 447$ mm

Air flow with swirl:

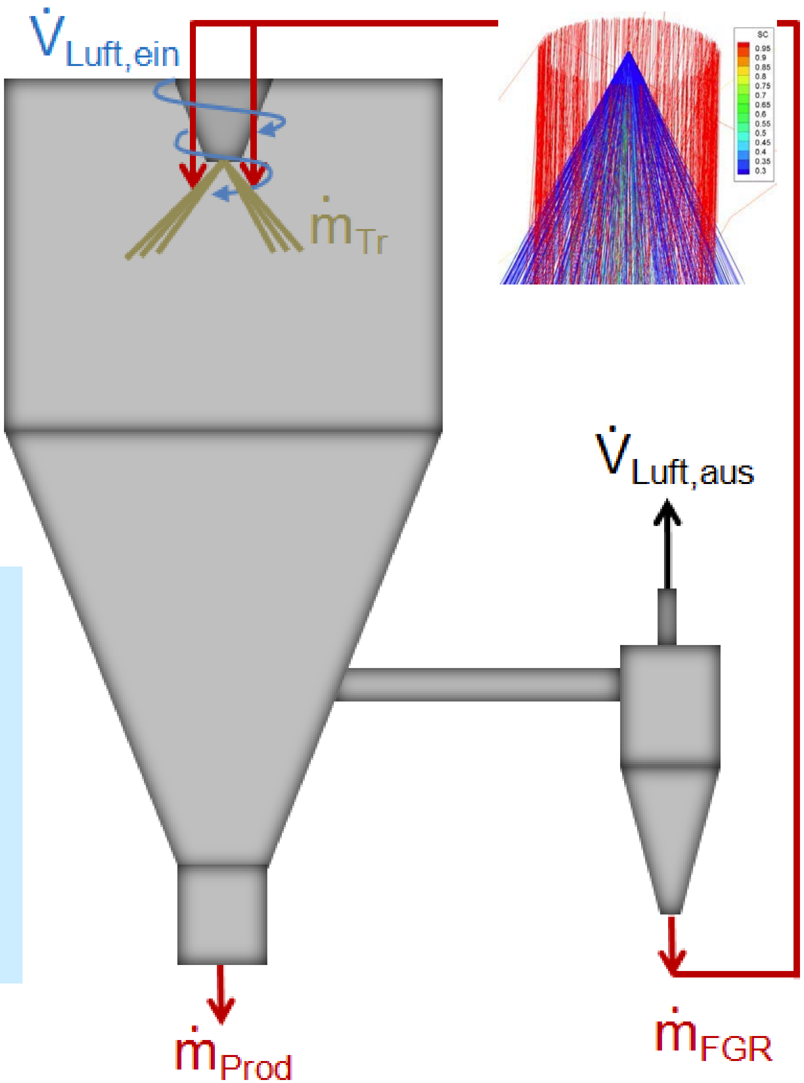
$\dot{m}_{air} = 1900$ kg/h

$\phi_{air} = 1.1$ mass-%

$T_{air} = 452.5$ K

$U_{ax} = 9.8$ m/s

$U_{tan} = 2.4$ m/s



Fines return:

Annular inlet around the nozzle

$D_o = 72$ mm

$D_i = 63$ mm

$U_{fine} = 37$ m/s

$\rho_{fine} = 440$ kg/m³

Pressure nozzle:

Hollow cone nozzle

$p_{nozzle} = 85$ bar

Spray angle $\beta = 52°$

$D_{nozzle} = 2$ mm

$H_{nozzle} = 270$ mm

Maltodextrine DE-18

Solution: 29 mass-% solids

$\rho_{drop} = 1090$ kg/m^3

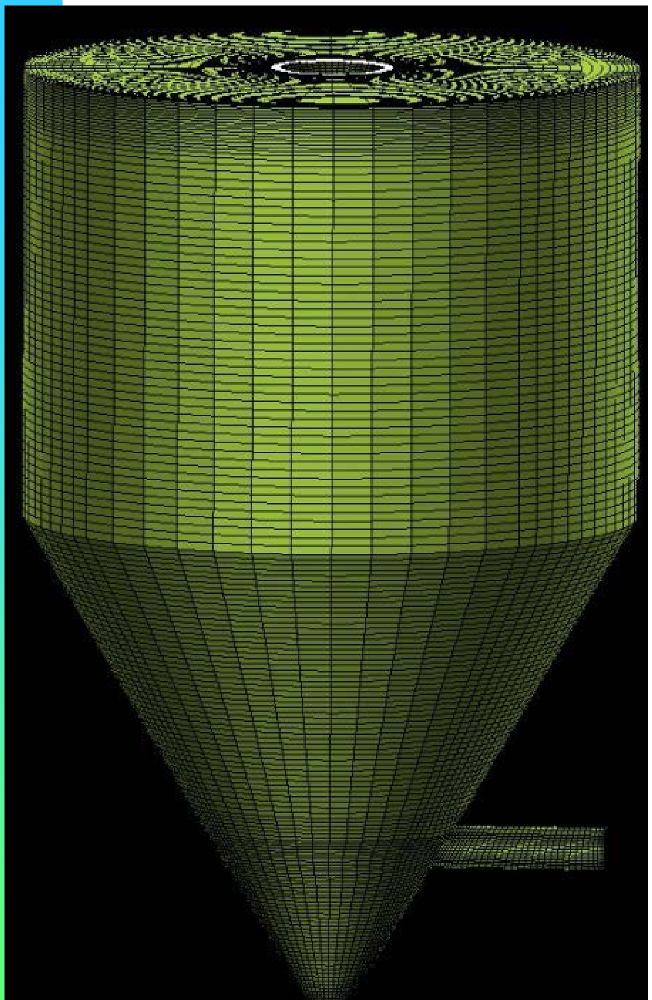
$\dot{m}_{solution} = 92$ kg/h

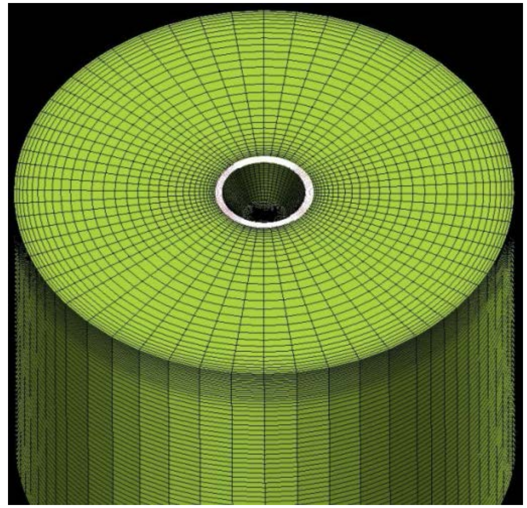
$T_{solution} = 293$ K

$U_{av} = 127$ m/s

Geometry of the Spray Dryer 2

➲ **Numerical discretisation and boundary conditions of the spray dryer:**



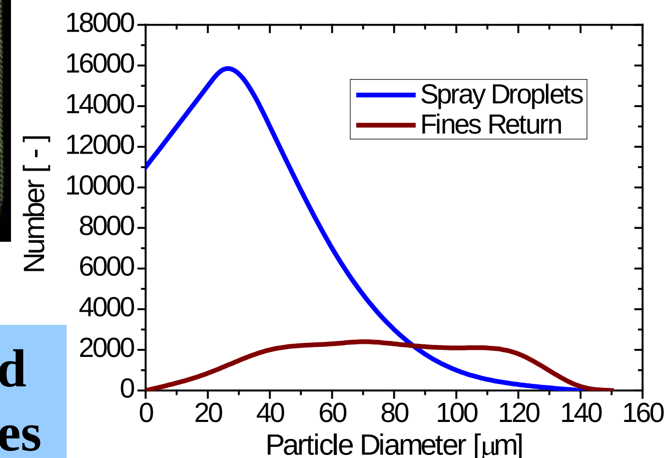


Discretisation:

138 blocks

586.564 meshes

Droplet and Particle Sizes



Inlet and boundary conditions:

Inlet: assumed velocity profiles

Walls: no-slip velocity

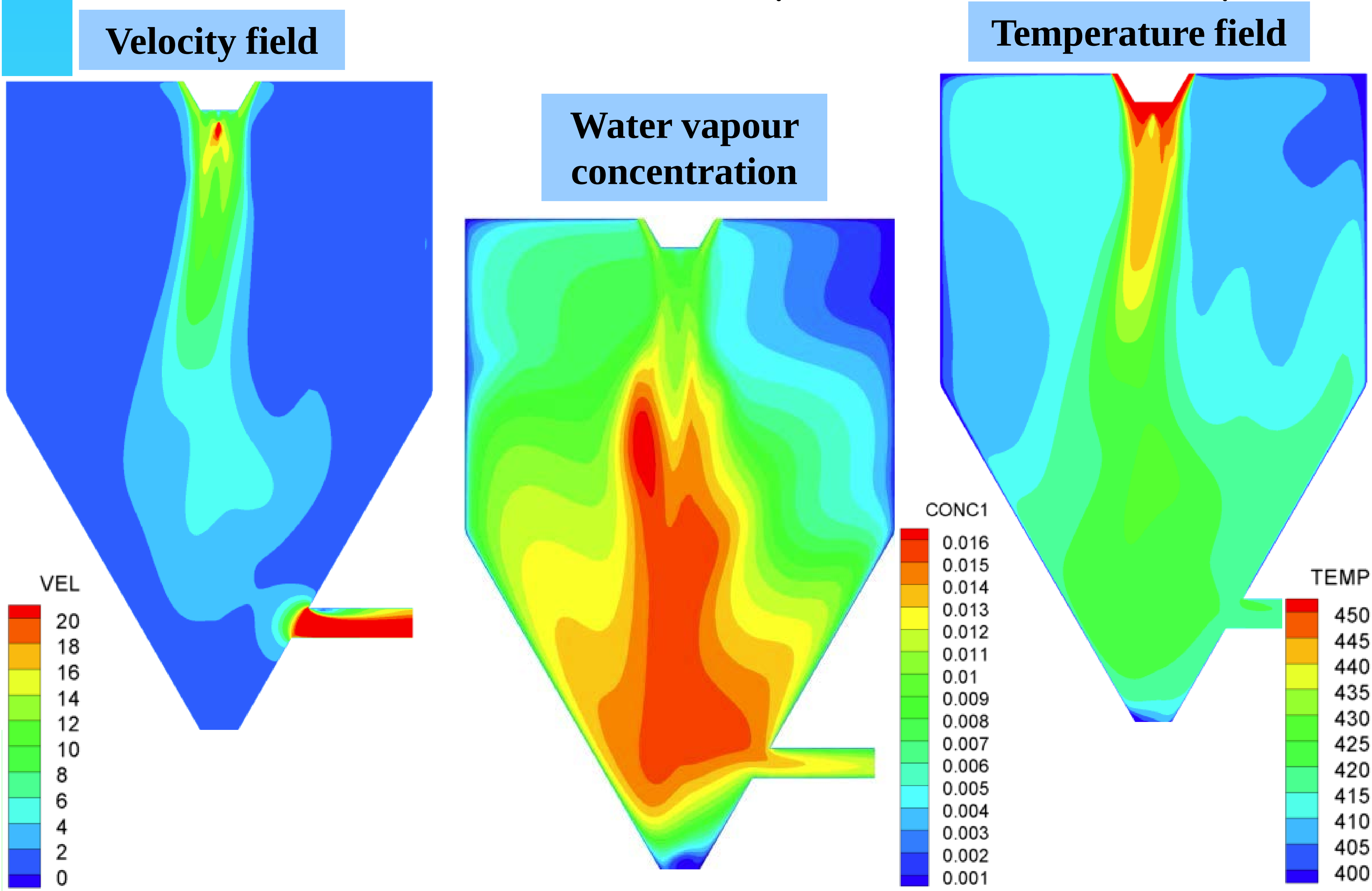
heat transfer coefficient ⇒ measurements

h = 10.5 W/(K·m²),

Outlet pipe: gradient free

Numerical Results Spray Dryer 1

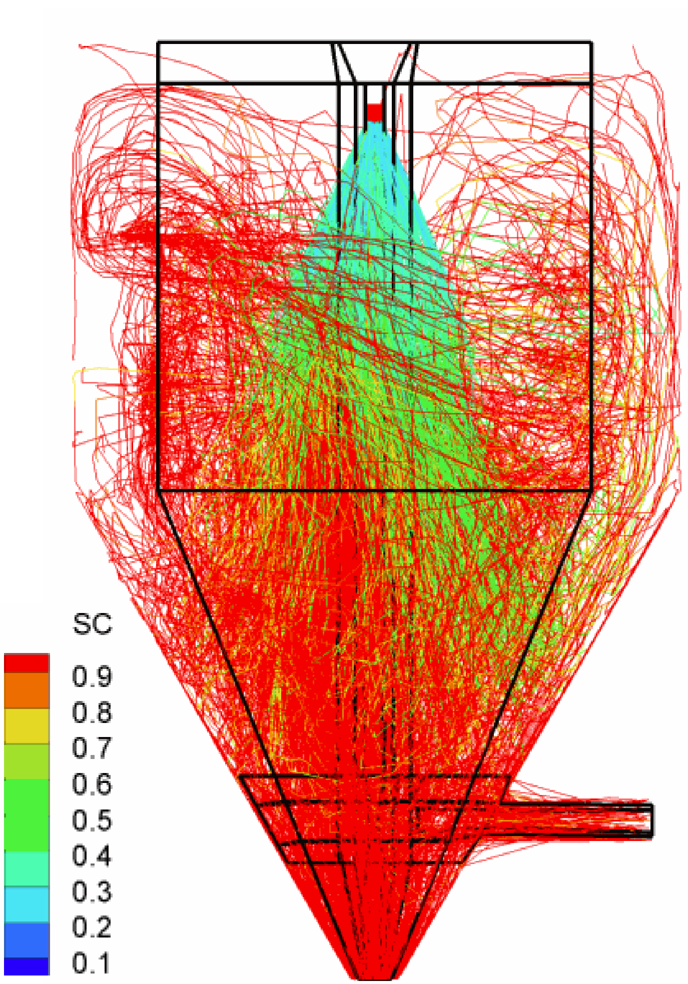
☞ Calculated flow structure and temperature field in the dryer:



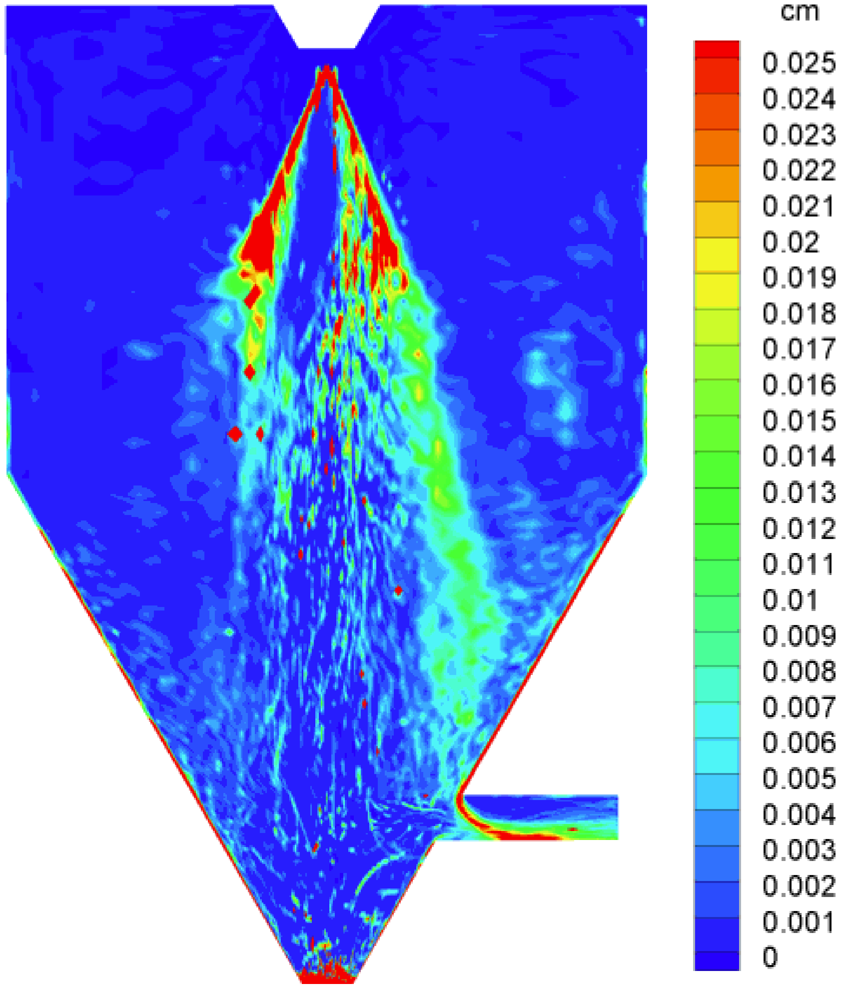
Numerical Results Spray Dryer 2

☞ **Particle phase properties throughout the spray dryer**

Particle trajectories



Particle concentration [kg/kg]



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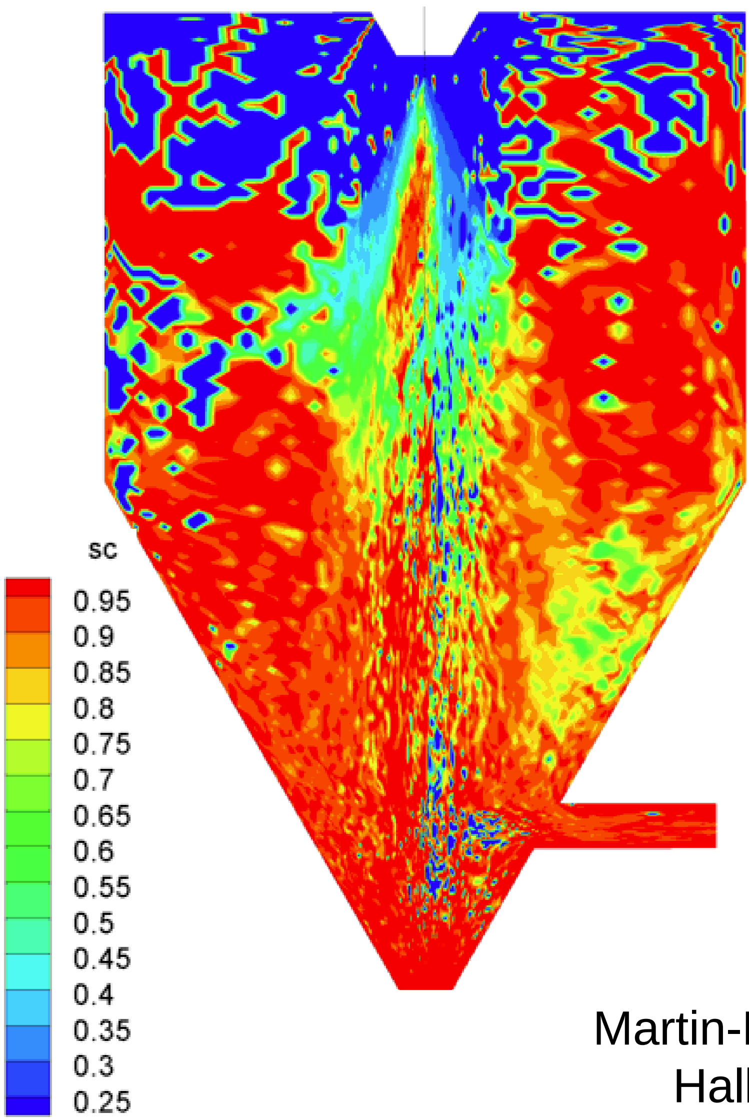
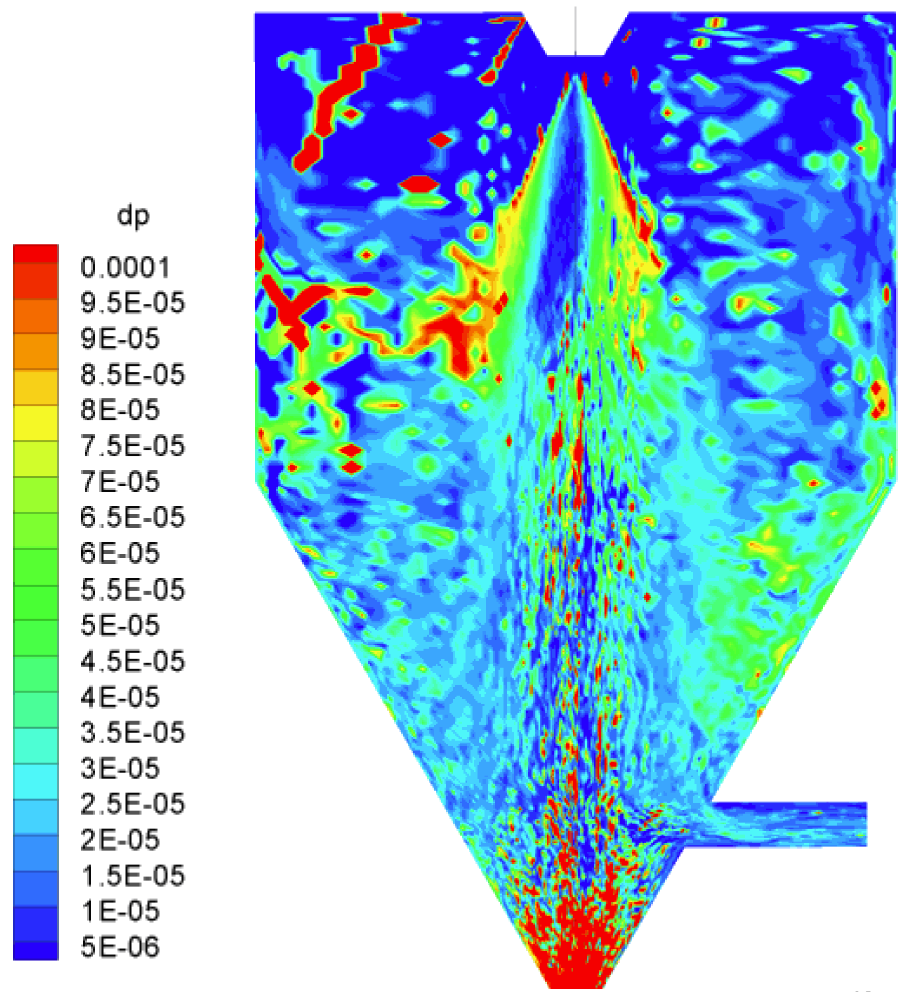
Numerical Results Spray Dryer 3

☞ **Particle-phase properties throughout the spray dryer**

Solids content in the particles



Local particle mean diameter



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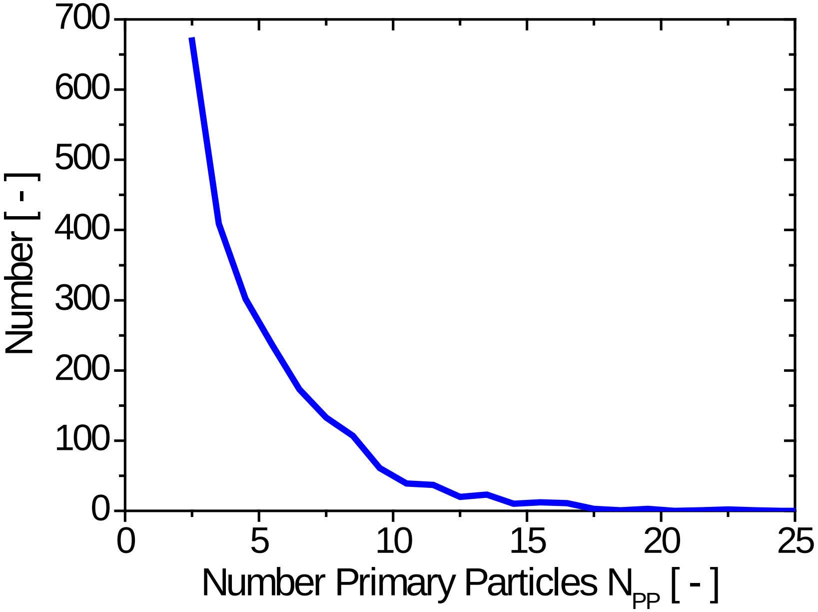
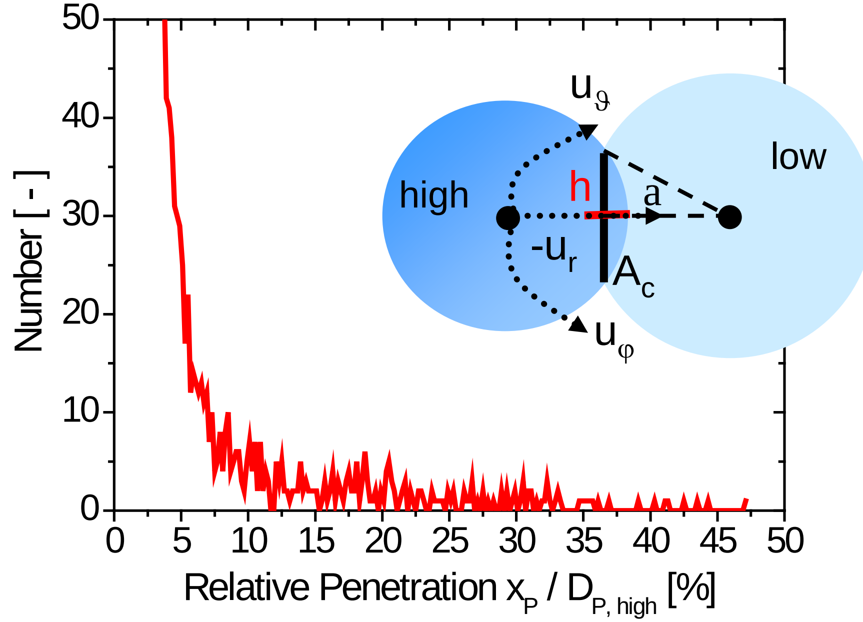
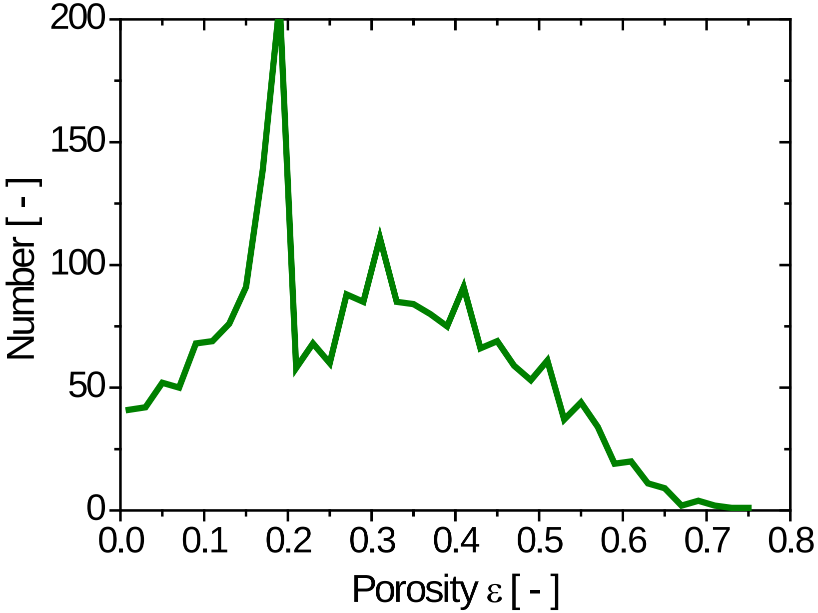
Numerical Results Spray Dryer 4

☞ Properties of the agglomerates produced in the spray dryer

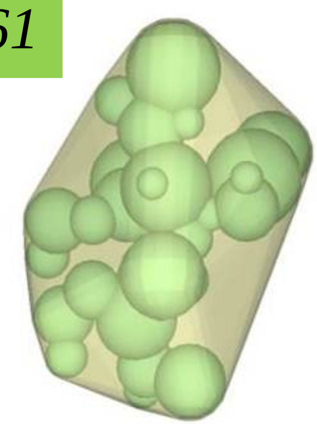






30 primary particles

$$\varepsilon_{hull} = 0.61$$

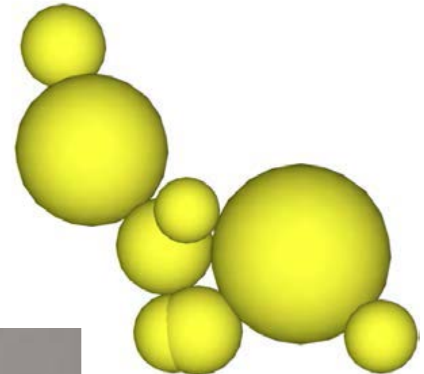


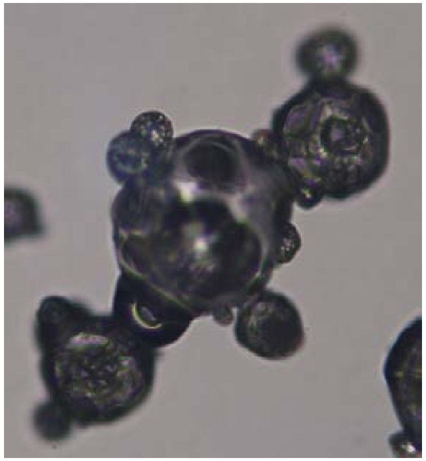
Porosity:

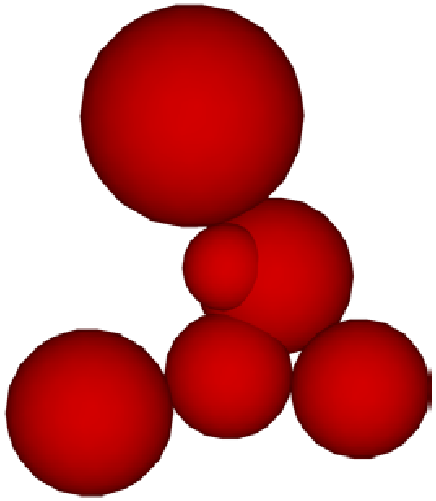
$$\varepsilon = 1 - \frac{V_{Part}}{V_{Hull}}$$

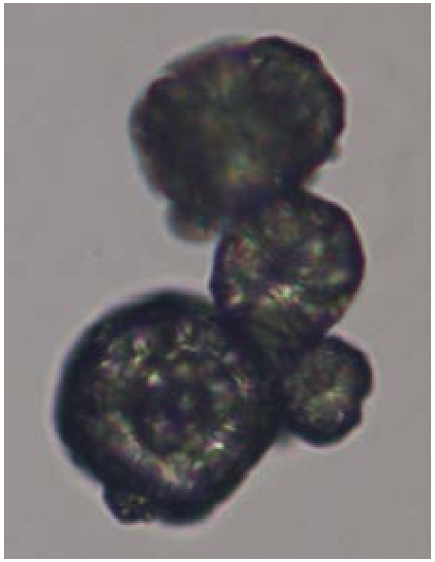
Numerical Results Spray Dryer 5

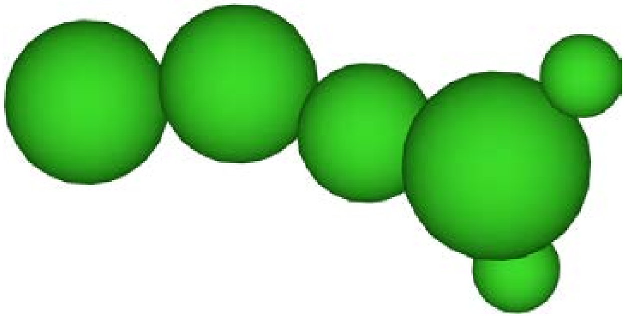
Simulated agglomerates compared with agglomerates collected from the spray dryer

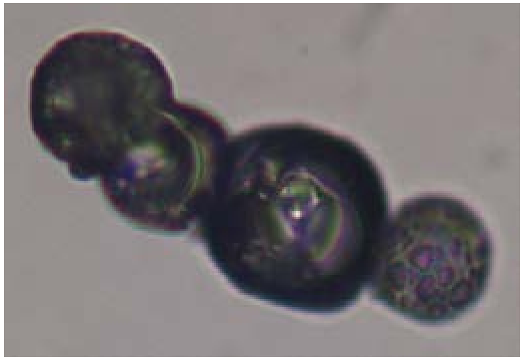






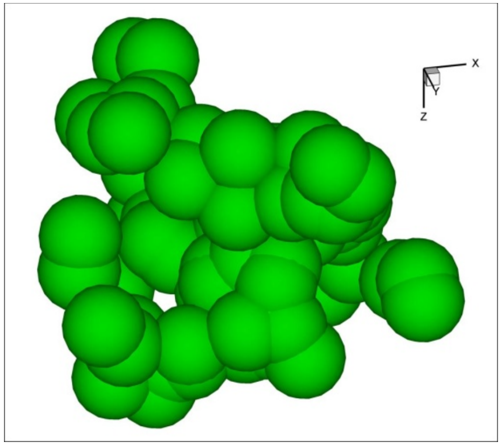
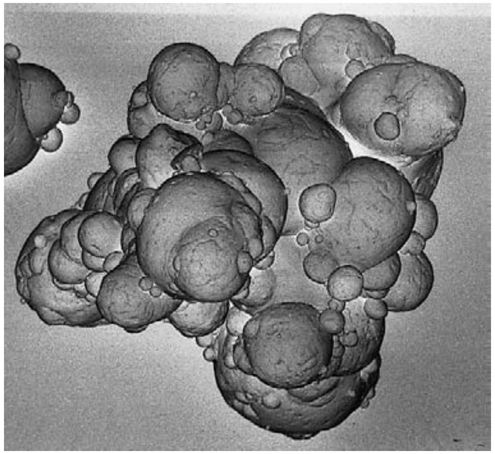






Conclusions / Outlook

- **Agglomeration of fine particles is an important elementary process in many industrial processes, among them is spray drying.**
- **The Euler/Lagrange approach allows for an effective coupling of fluid dynamics and particle transport, collision and agglomeration**
- **A Lagrangian agglomeration structure model was developed, which provides porosity, sphericity, surface area and fractal dimension of agglomerates**
- **The model calculations provided consequential results for dry particles and mono-viscous particles with penetration in HIT.**

- **First calculations of a spray dryer showed reasonable agreement with very limited measurements (NIRO Copenhagen).**
- **A further validation of the model will be based on experiments in a model dryer with two interacting fan sprays.**

Publications

- Blei, S. and Sommerfeld, M.: CFD in Drying Technology-Spray Drying Simulation. Modern Drying Technology: Volume 1 Computational Tools at Different Scales (Eds. E. Tsotsas and A. S. Majumdar), WILEY-VCH, Weinheim, 155 – 208 (2007).
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- Sommerfeld, M. and Ho, C.A.: Numerical calculation of particle transport in turbulent wall bounded flows. Powder Technology, Vol. 131, 1-6 (2003)
- Sommerfeld, M., van Wachem, B. & Oliemans, R.: Best Practice Guidelines for Computational Fluid Dynamics of Dispersed Multiphase Flows. ERCOFTAC, ISBN 978-91-633-3564-8 (2008).
- Sommerfeld, M.: Modelling particle collisions and agglomeration in gas-particle flows. CD-ROM Proceedings 7th Int. Conf. on Multiphase Flow, Tampa, FL USA, May 30. – June 4. (2010)
- Sommerfeld, M. and Stübing, S.: Lagrangian modeling of agglomeration for applications to spray drying. 9th International ERCOFTAC Symposium on Engineering Turbulence Modeling and Measurements, Thessaloniki, Greece, 6. – 8. June 2012
- Stübing, S., Dietzel, M. and Sommerfeld, M.: Modelling agglomeration and the fluid dynamic behaviour of agglomerates. Proceedings of ASME-JSME-KSME Joint Fluid Engineering Conference 2011 (AJK2011-FED) July 2011, Hamamatsu, Shizuoka, Japan, Paper No. AJK2011-12025.

ANNOUNCEMENT

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14th Workshop on Two-Phase Flow Predictions

07. – 10. September 2015

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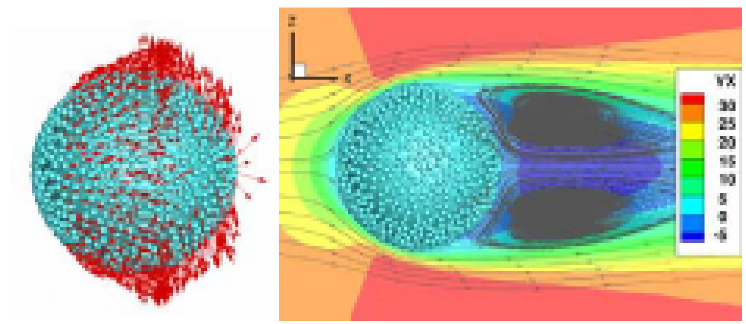

Lattice-Boltzmann Simulations: Flow about a particle coated with 882 drug particles at Re = 200, study related to drug particle detachment in an inhaler.