Nanoparticle Diagnostics Collaborative Task

Subtask 3.4 A: Development of laser diagnostics techniques for combustion systems

Three-angle scattering and laser extinction for soot characterization in premixed flames

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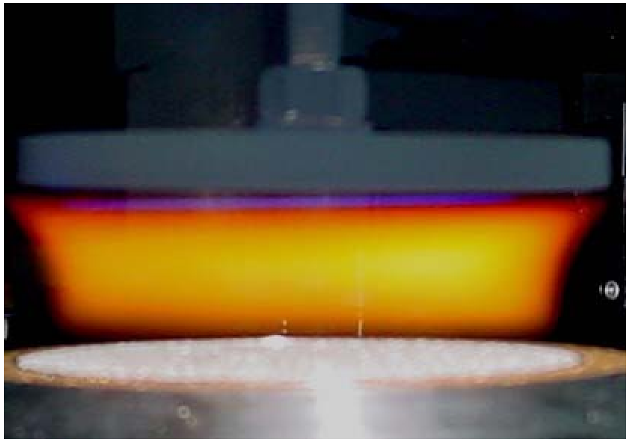
Aim of the work

- ✓ To combine different optical diagnostic techniques in order to have a larger spectrum of data for comparison
- ✓ To characterize the premixed flame in terms of soot parameters (e.g. f_v, d_p)

✓ Extinction	⇨	f_v
+Three-angle scattering	⇨	R_g, d_p
✓ TEM	⇨	D_f, K_f, R_g, d_p

Experimental Apparatus: McKenna burner (premixed flames)



Bronze porous plug - Φ = 60 mm

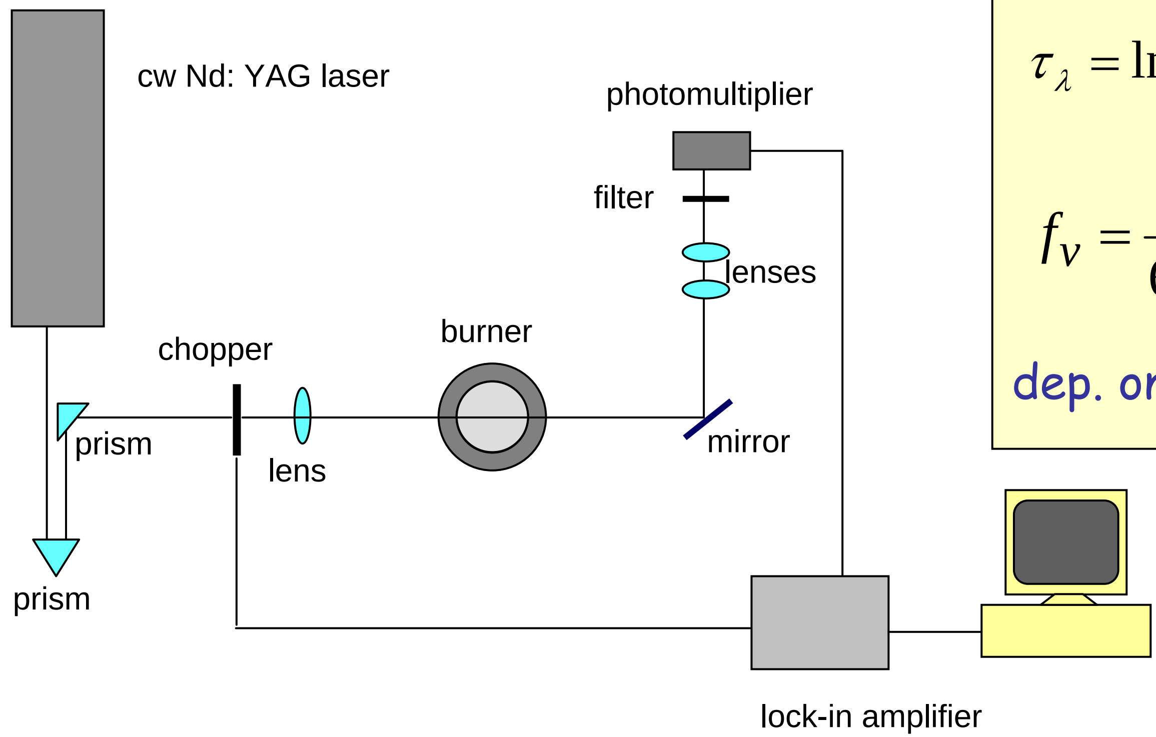
Shroud ring - th = 6 mm

Stab. plate: Φ = 60 mm, HAB=20 mm

Premixed C_2H_4/air flame :

Φ=2.34, C/0=0.77, total flow rate =10 Nl/min

Extinction



$$\tau_\lambda = \ln\left(\frac{I_L}{I_0}\right) = -K_{ext}L$$

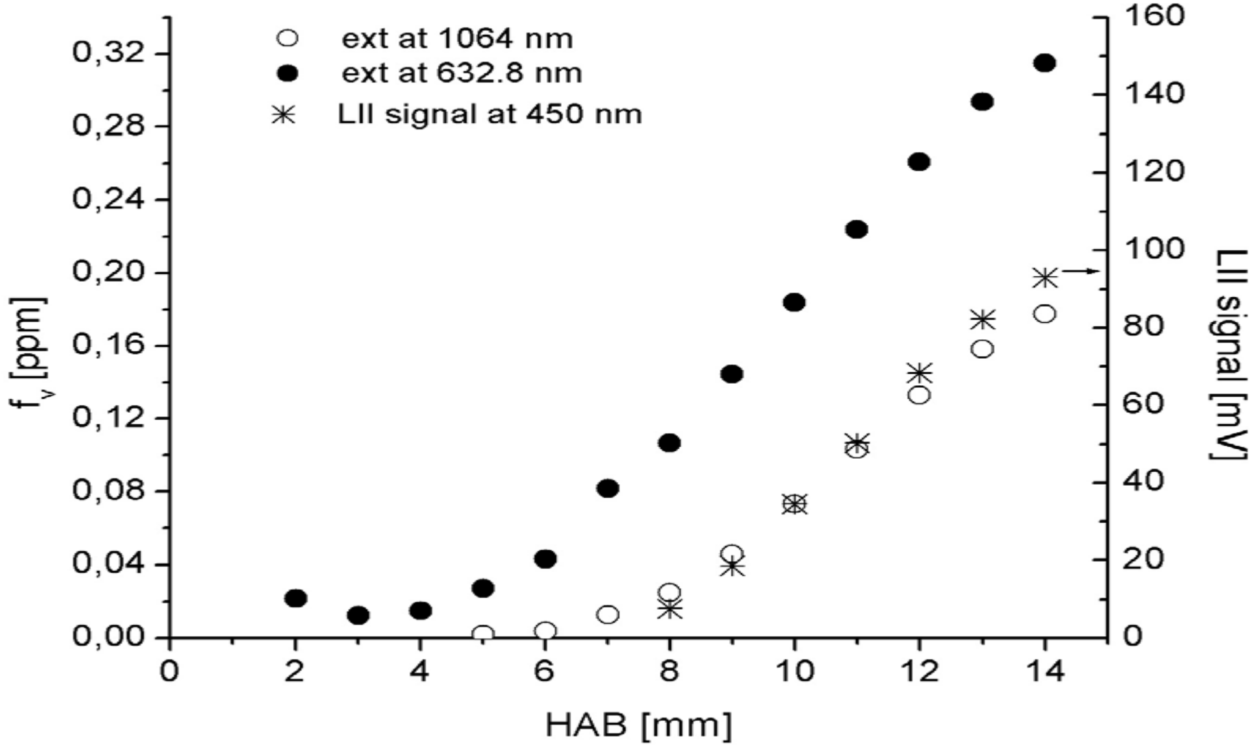
$$f_v = \frac{\lambda K_{ext}}{6\pi E(m)} = \frac{\pi}{6} d_p^3 N_p$$

dep. on the refractive index

✓ **For extinction: cw Nd:YAG laser λ=1064 nm**

(Attention to the wavelength used has to be payed because of possible contribution of other gas-phase species to absorption)

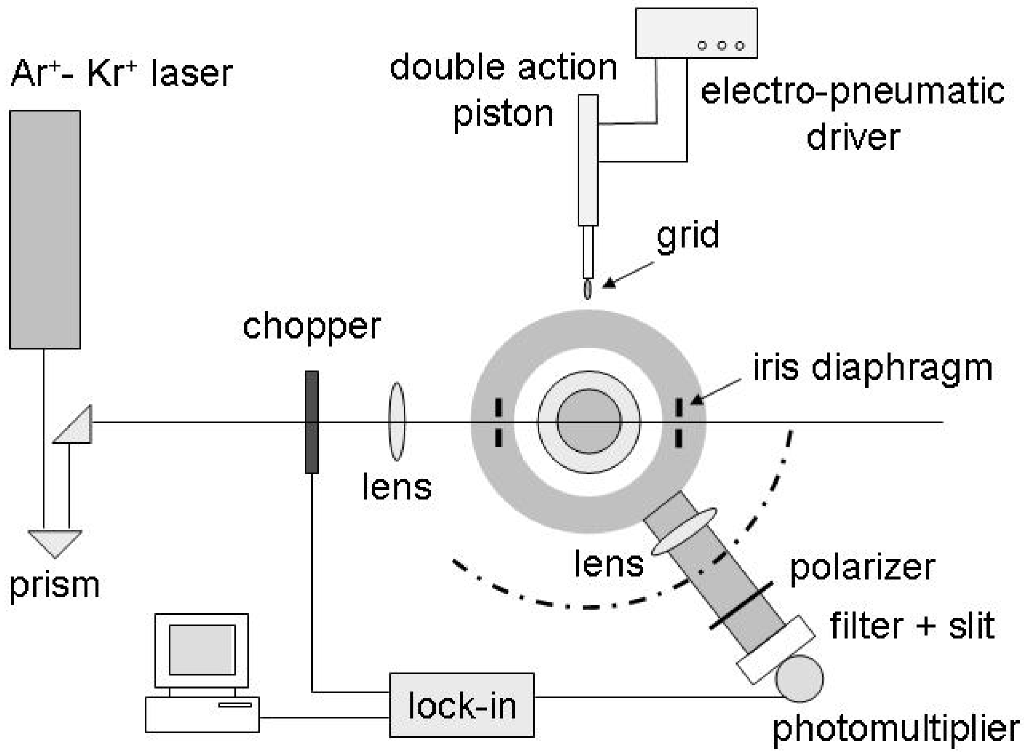
Results – Extinction (632.8 nm, 1064 nm) - LII



✓ **Wavelength for extinction must be carefully selected**

(Both techniques work, but LII gives local value of f_v)

Multi-angle scattering/TEM



- For scattering Ar+-Kr+ laser (λ=514 nm, 200 mW)
- Signal detected at 30°-90°-150°

Three-angle scattering

- Signals collected at 30°, 90°, 150°
- Correction for the probe volume variation & calibration
- Numerical approach to evaluate R_{gm1} (average radius of gyration) from $R_{vv}(30°/150°)$ (Fractal + RDG theory)
- From signal at 90° + absorption data ⇨ D_{30}
- By knowing R_{gm1} and D_{30} ⇨ d_p

Used parameters:

✓ Structure factor from Lin relationship (Sorensen et al. 1999)

✓ PDF for N_a (Lognormal, $\sigma = 2.1$)

✓ D_f and K_f appearing in Fractal-like approach from TEM

Scattering from fractal aggregates (1)

Rayleigh scattering of a single particle

$$C_{VV}^{p} = k^{4} a^{6} F(m) \qquad k = 2\pi / \lambda \qquad F(m) = \left| \frac{m^{2}-1}{m^{2}+1} \right|^{2}$$

Fractal aggregates

$$N = k_{f} (R_{g} / d_{p})^{D_{f}}$$

$$C_{VV}^{a} = C_{VV}^{p} N^{2} S(qR_{g}) \qquad q = \frac{4\pi}{\lambda} \sin\theta / 2$$

$$S(qR_{g}) \xrightarrow{qR_{g} \leq 1} \text{Guinier}$$

$$S(qR_{g}) \xrightarrow{qR_{g} > 1} \text{Power law}$$

$$S(qR_{g}) = \left[1 + \sum_{s=1}^{4} C_{s} (qR_{g})^{2s} \right]^{-D_{f}/8}$$

(Lin relationship)

Scattering from a **single** fractal aggregate

$$I_{VV}^{a}(\theta) = \eta I_{0} C_{vv}^{p} N^{2} S(qR_{g})$$

Scattering from fractal aggregates (2)

Scattering from a polydisperse distribution of fractal aggregates

$$I_{VV}^{p}(\theta) = \eta I_0 C_{VV}^{p} N_a \int N^2 S(qR_g) p(N) dN = \eta I_0 C_{vv}^{p} N_a \overline{m_2} \, \overline{S(qR_g)}$$

$$S^*(q\overline{R_g}) = \overline{S(qR_g)}$$

$$\overline{R_{gs}} = R_{gm1} = d_p (\overline{m_1} / k_f)^{(1/D_f)}$$

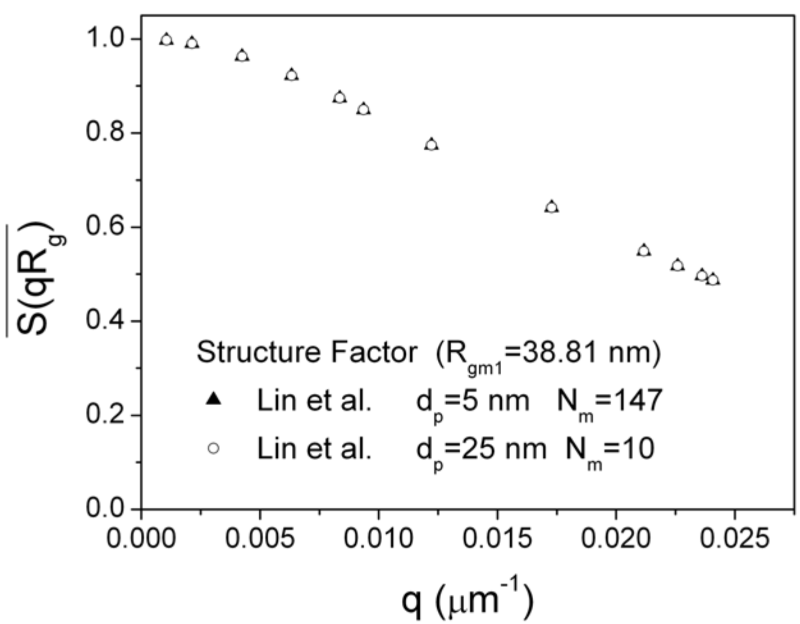
$p(N)dN = \log \ normal$

$D_f = 1.67$

$k_f = 6.34$

$\sigma = 2.1$

From TEM



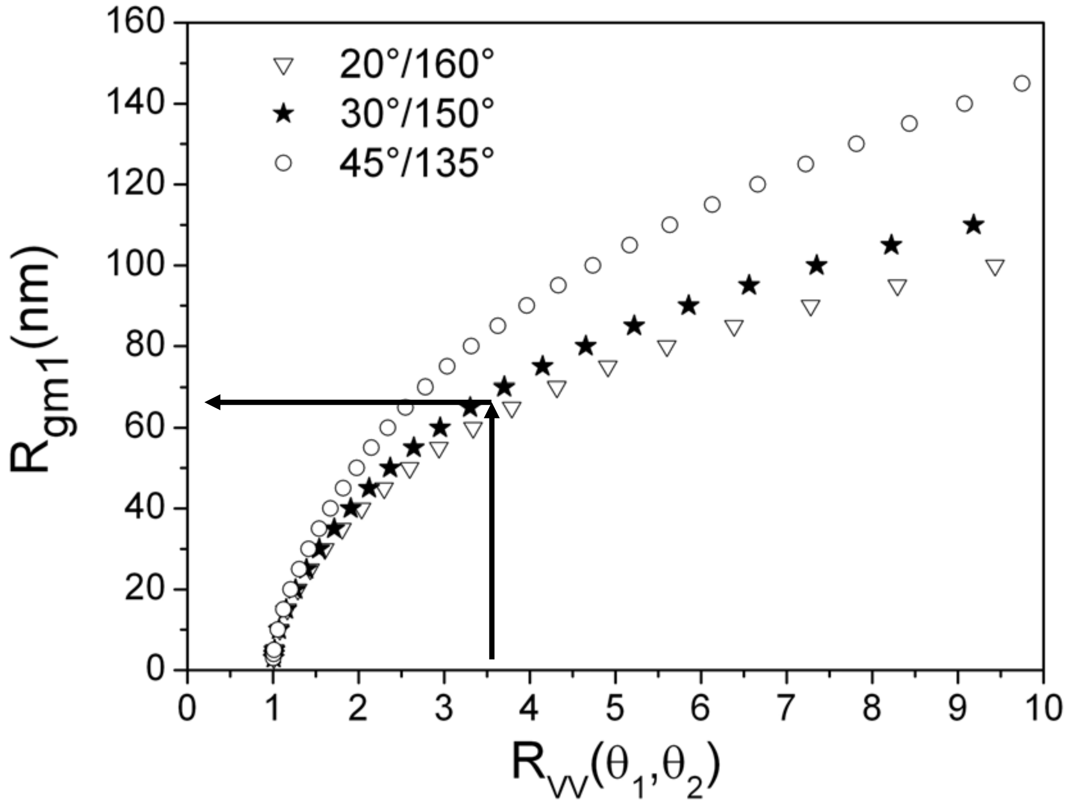
$$S^*(qR_{gm1}) = \left[1 + \sum_{s=1}^{4} P_s(qR_{gm1})\right]^{-1}$$

$$I_{VV}^{p}(\theta) = \eta I_0 C_{VV}^{p} N_a \overline{m_2} S^*(qR_{gm1})$$

Scattering from fractal aggregates (3)

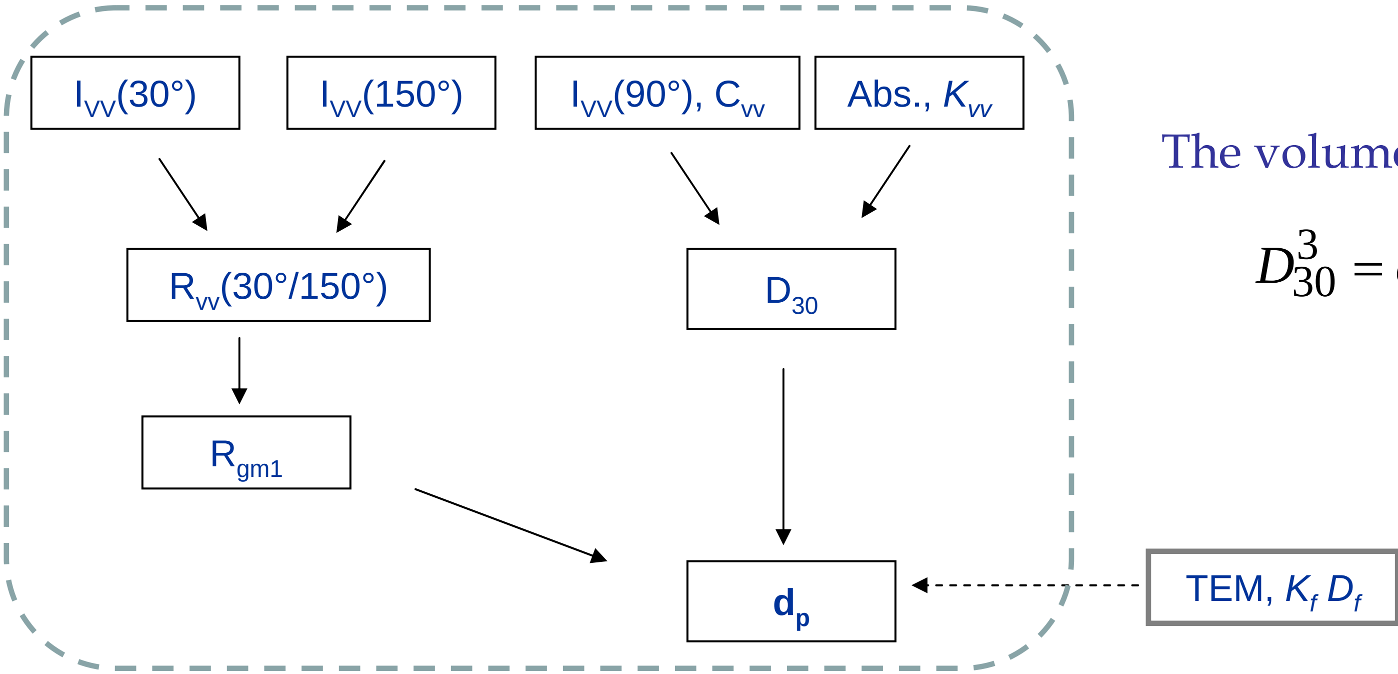
Dissymmetry ratio

$$R_{VV}(\theta_1,\theta_2)=\frac{S^*(q_1R_{gm1})}{S^*(q_2R_{gm1})}$$



Summarizing

Three-angle scattering/extinction



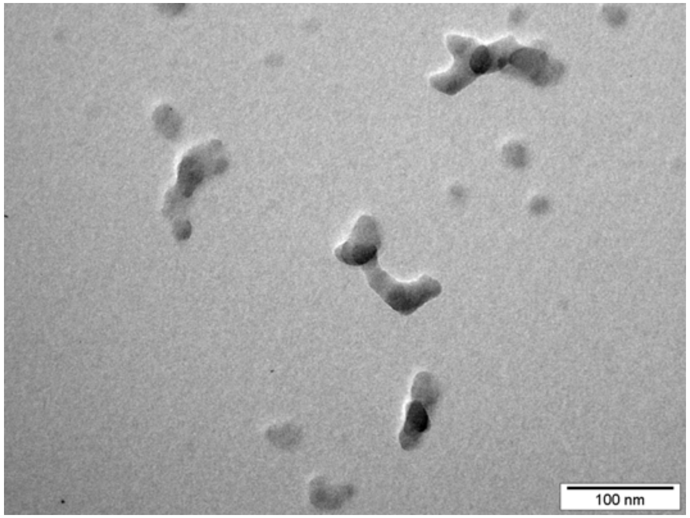
The volume mean diameter

$$D_{30}^3 = d_p^3 \overline{m_1} = d_p^3 \frac{N_p}{N_a}$$

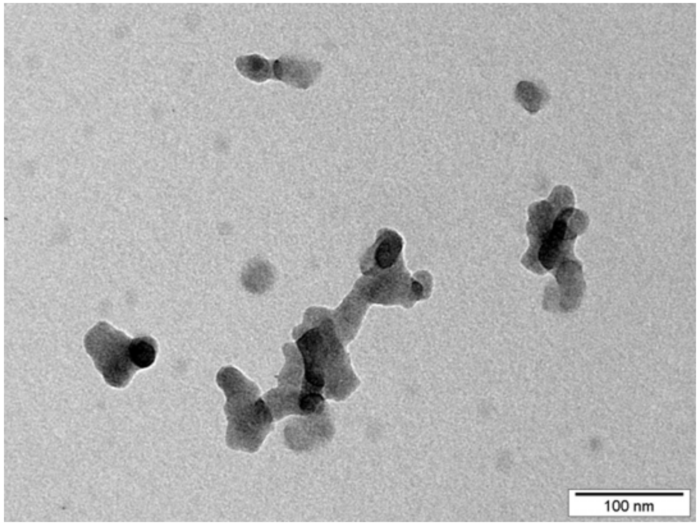
TEM analysis:
- D_f, K_f
- **Validation of d_p measurements**

TEM analysis

HAB=12 mm



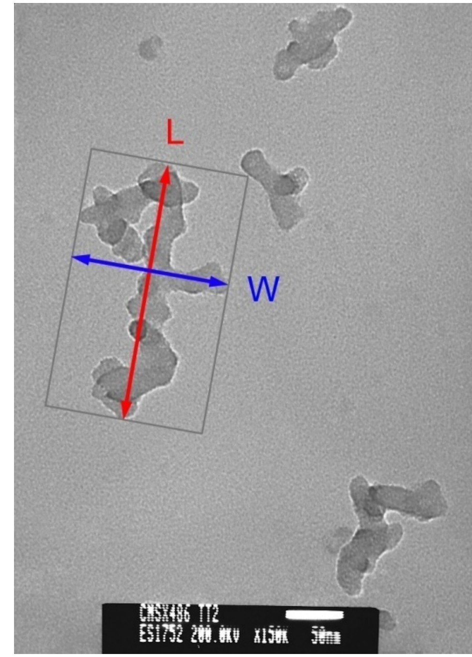
HAB=14 mm



- ✓ Primary particles diameter: statistics on 300 samples indicates a lognormal distribution with σ=1.1 ⇨ monodisperse

- ✓ For fractal analysis 300 aggregates are considered

TEM analysis



- ✓ For each aggregate, the geometric dimension L and W are measured

- ✓ Calculus of aggregate projected area and the radius, linked as:

$$N = k_a \left(\frac{A_a}{A_p} \right)^{\alpha} = k_a \left(\frac{R_a}{a} \right)^{2\alpha}$$

- ✓ Ln N versus ln R_L gives D_f from the slope

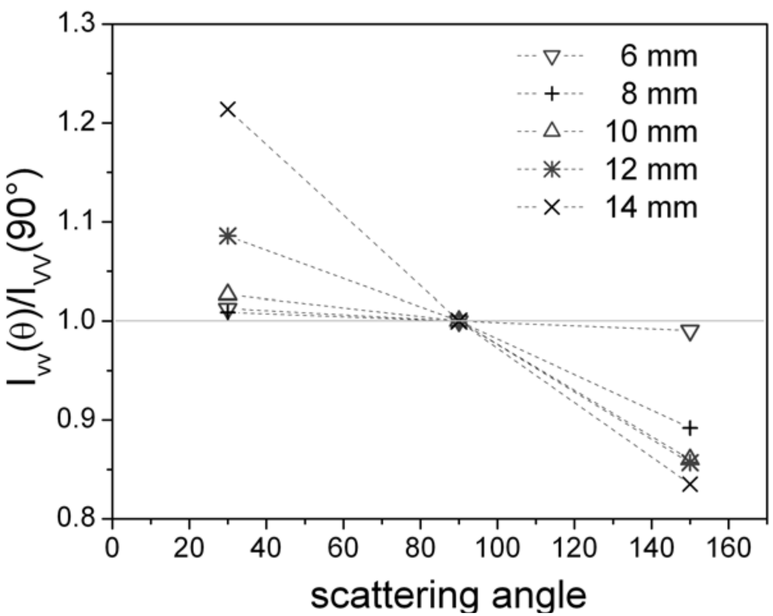
$$N = k_L \left(\frac{R_L}{a} \right)^{D_f}$$

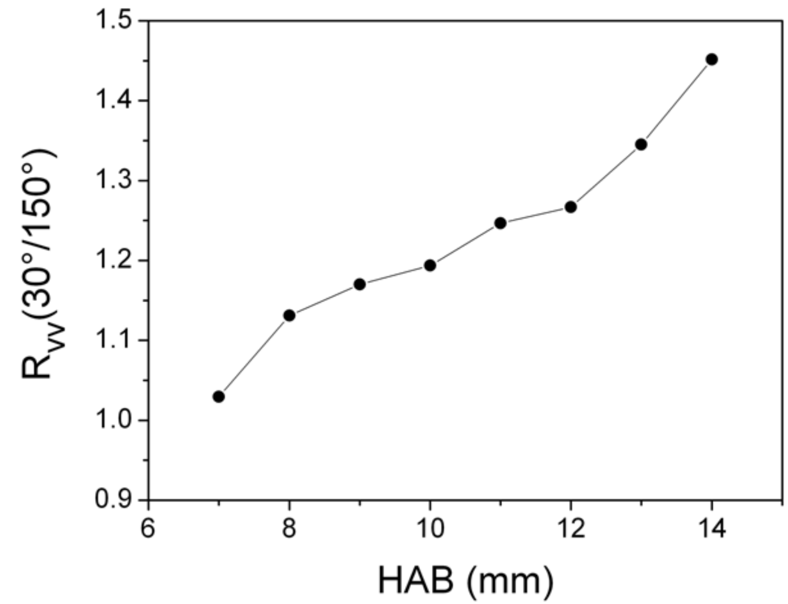
- ✓ Finally R_g and K_f can be derived from the following relationships:

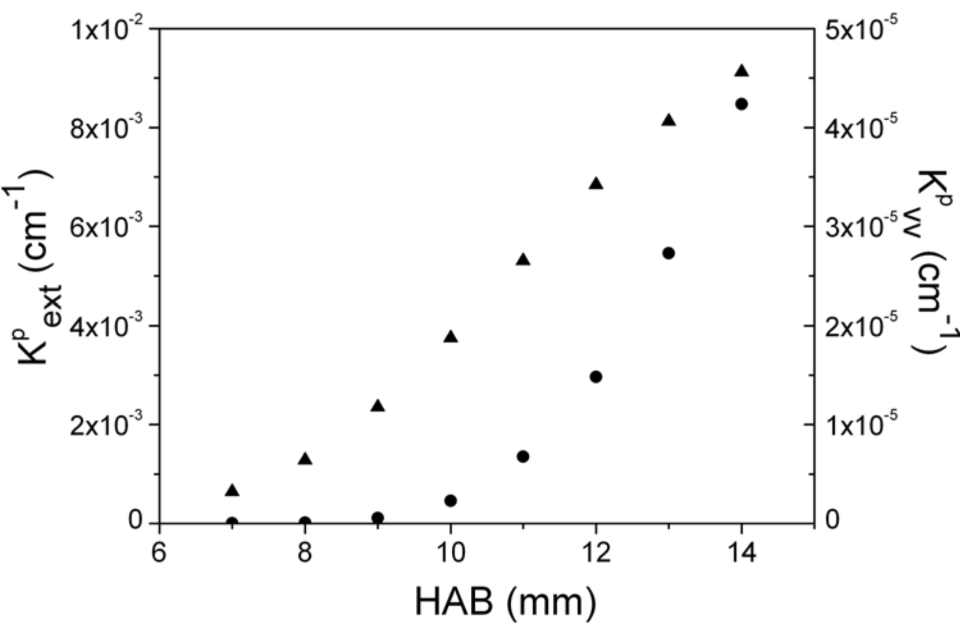
$$\frac{k_f}{k_L} = 2^{D_f} \left(\frac{R_L}{R_g} \right)^{D_f} \qquad \left(\frac{R_L}{R_g} \right) = \left[\frac{(D_f + 2)(D_f + 5)}{2D_f (D_f + 1)} \right]^{1/2}$$

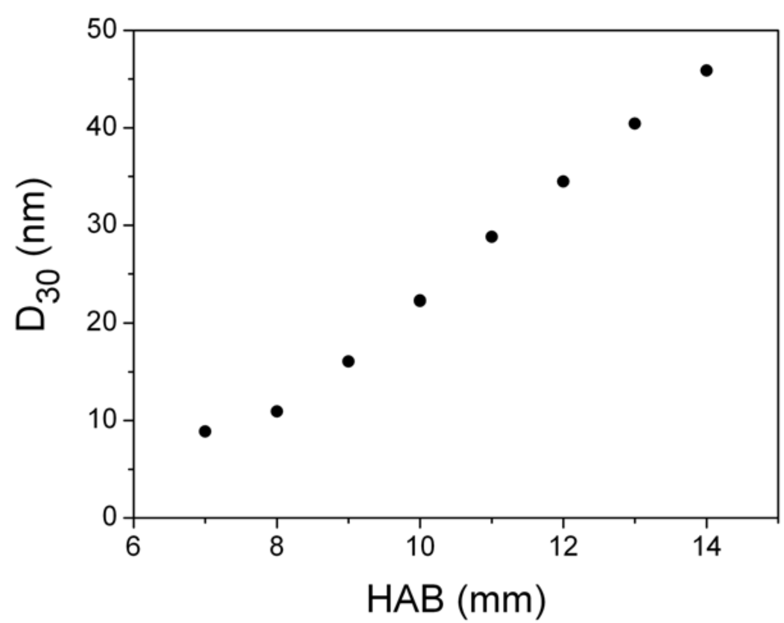
D_f= 1.67 K_f =6.34

Experimental Results

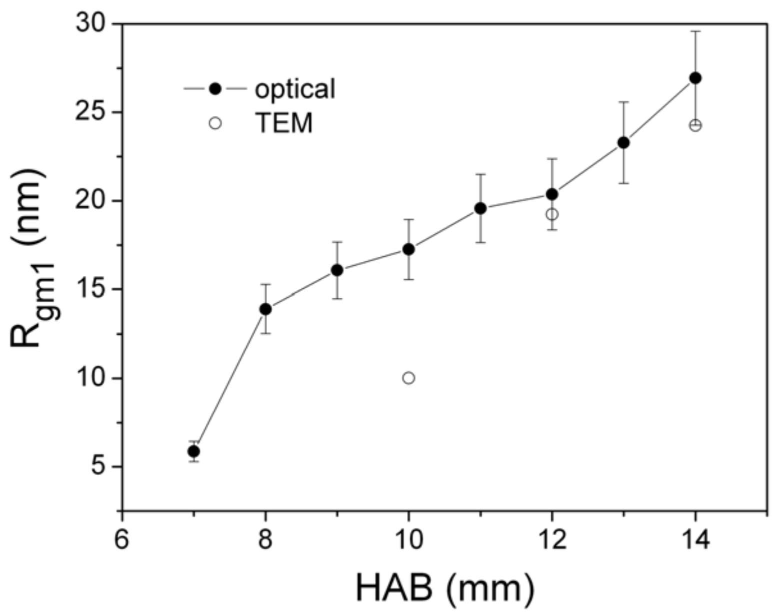


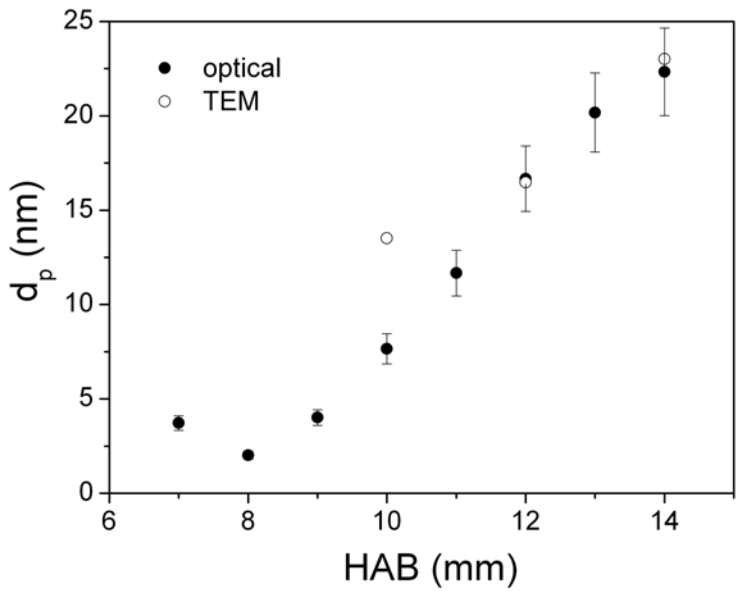






TEM/Scattering Measurements

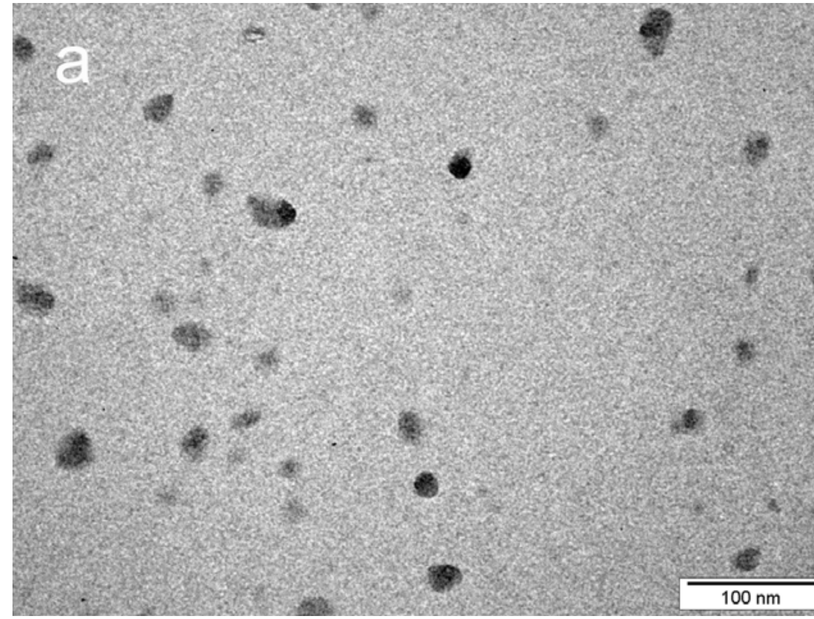


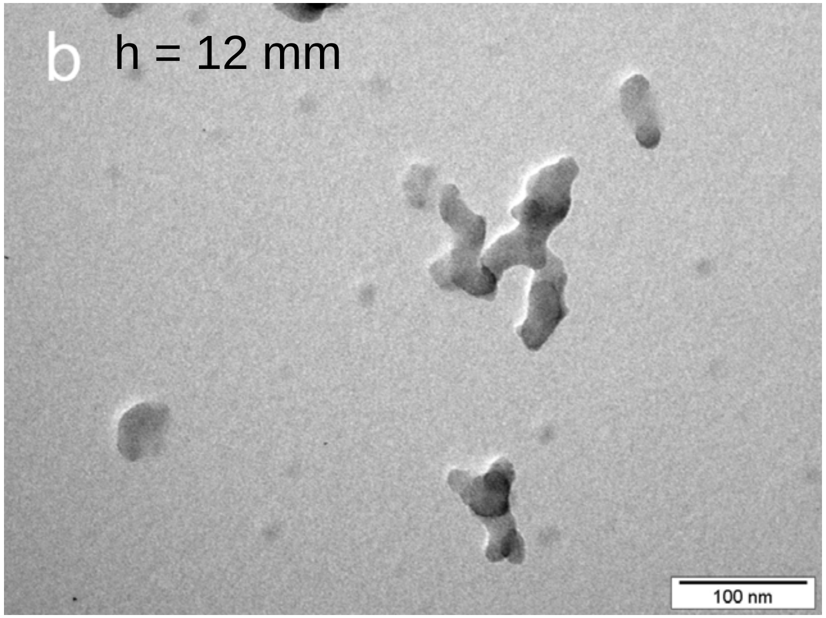


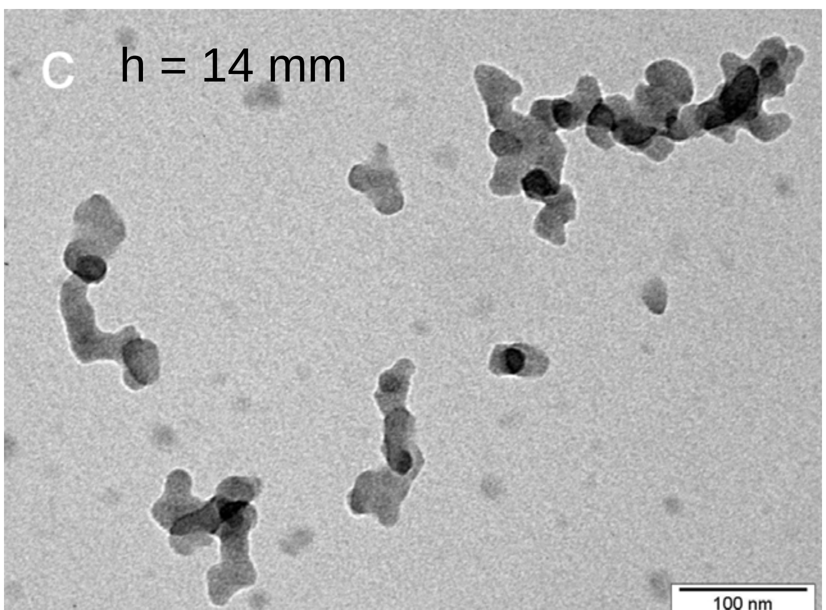
HAB	d_p (nm)		R_g (nm)	
	Optical	TEM	Optical	TEM
12 mm	17.90±1.86	16.47	19.63±1.94	19.23
14 mm	22.95±2.38	23.01	27.19±2.68	24.26

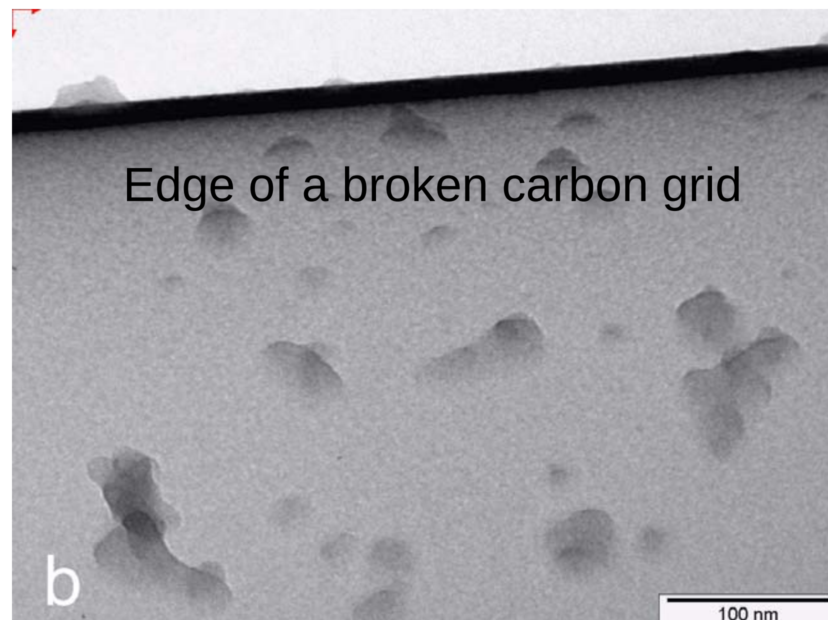
TEM images

h = 10 mm









Conclusions

✓A function for structure factor of a polydisperse distribution was determined in terms of an average radius of gyration, R_{gm1}

✓The coupling of IR laser extinction with three-angle scattering measurements allows to obtain primary particle measurements in agreement with TEM results.

✓f_v from LII is in agreement with IR extinction measurements. Care must be taken with extinction measurements at other wavelengths

✓At low heights soot appears to be mainly composed by liquid like structures

Future work: soot sizing will be carried out at low laser fluences.

Thank you for your attention