Fundamental Studies of Nanoparticle Capture Using Flow-through Monoliths

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- Efficiency of size-selected particle deposition in monoliths at flow rates controlled to reflect typical mass transfer conditions within DPF wall
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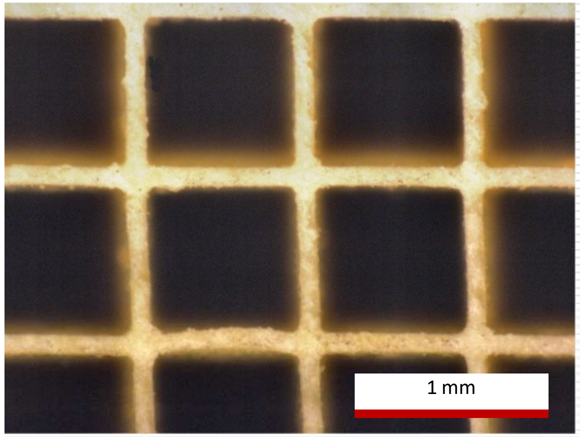
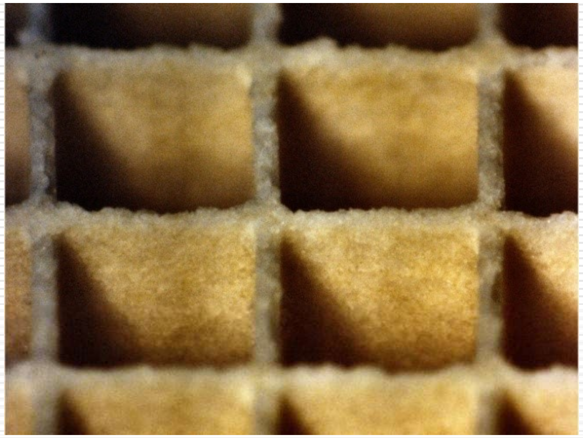
  - Effect of altering aerosol charge state
- Loading of monoliths on diesel burner (to simulate particle loading in light-duty engine exhaust)
  - Pressure drop and mass deposition efficiency
  - Images of bridging across channels
- Conclusions and acknowledgments

Use of bare cordierite flow-through monolith

Uniform array of channels for fundamental studies of particle deposition mechanisms

Portions of DOC leading edge commonly caked up in real-world operation





Two concurrent studies:

1. Size dependence of particle deposition mechanisms in laminar flow relevant to capture in DPF wall
2. Measurements of diesel particle deposits at typical engine exhaust flow rates and observations of channel bridging

Diffusion of particles from laminar flow in single channel: dimensionless deposition parameter ϕ

$$\phi = \frac{D_p L}{Q} = \frac{D_p}{{d_h}^2} \cdot \frac{L}{U} = \tau_C / \tau_D$$

D_p – particle mass diffusivity
L – channel length
Q – volume flow rate
d_h – hydraulic diameter
U – average flow velocity

τ_C and τ_D are time constants for convective and diffusive particle transport

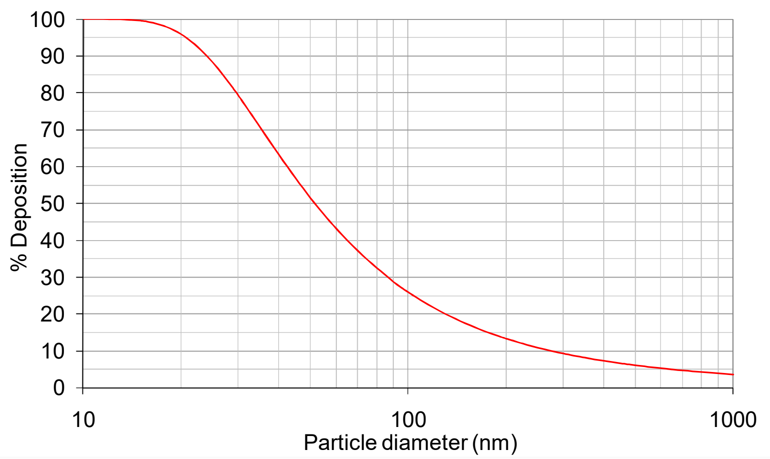
Numerical solutions in the literature for particle penetration, *P* (= outlet / inlet particle no. concentration), comprise two asymptotic expressions that apply either side of a threshold value of ϕ (denoted ϕ_0):

$$P = 1 - \sum_{i=1}^{\infty} \alpha_i \phi^{(i+1)/3} \quad \text{for } \phi < \phi_0$$

$$P = \sum_{i=1}^{\infty} \beta_i \, exp(-\gamma_i \phi) \quad \text{for } \phi > \phi_0$$

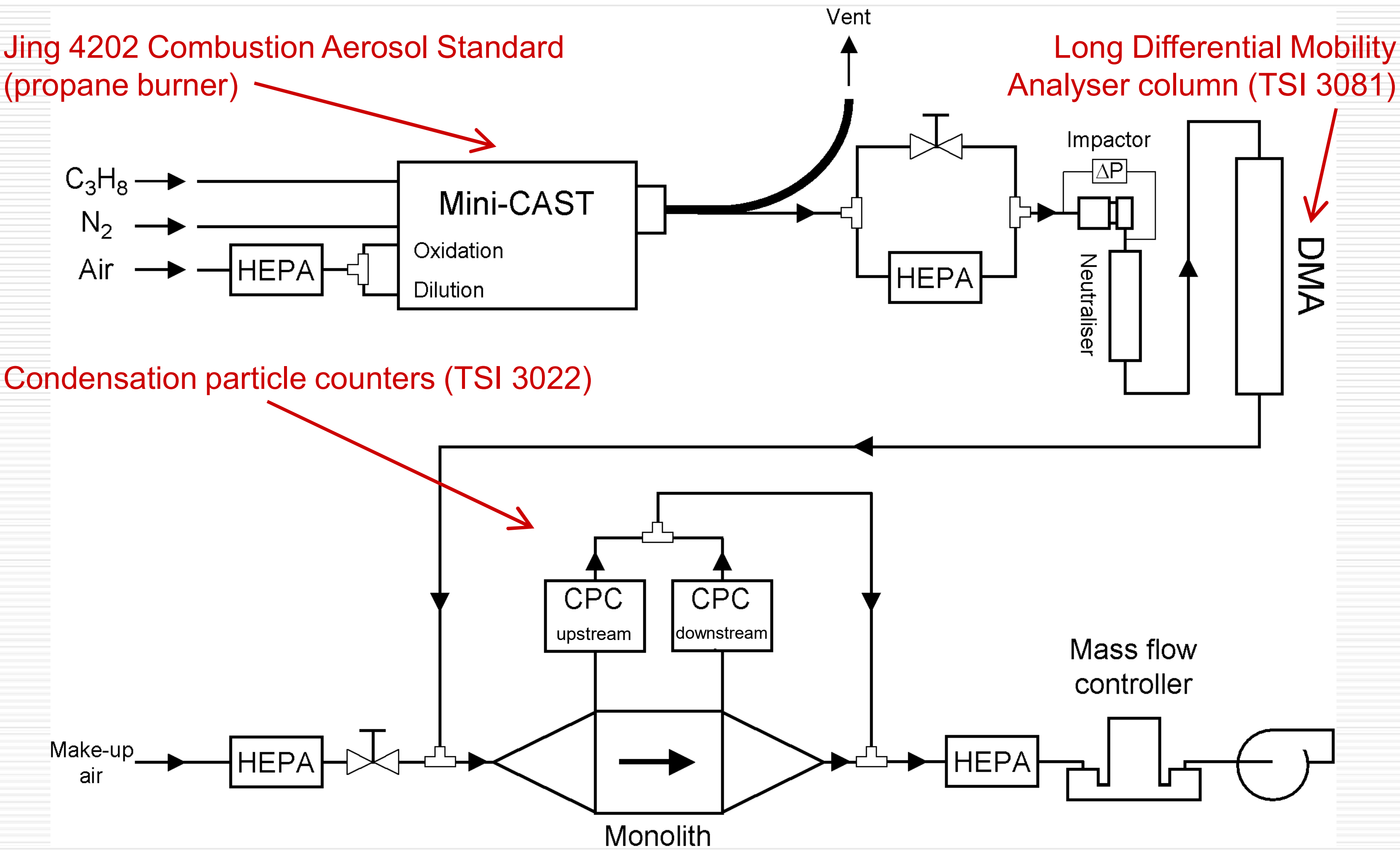
Numerical calculations for ϕ_0 and the first two values for coeffs α_i and β_i and eigenvalues γ_i for traverse diffusion in square channels (refs: J.Aerosol Sci.14 (1983) 741-745 and Q.Appl.Math. 17 (1959) 285-297):

ϕ_0	i	α_i	β_i	γ_i
1.50 X 10^{-2}	1	6.214	0.804	11.91
	2	-5.984	0.104	71.07



◄ Deposition efficiency versus particle diameter calculated for U = 1 cm/s (Re = 0.65) in 600 cpsi, ϕ4.66” by 6” flow-through monolith

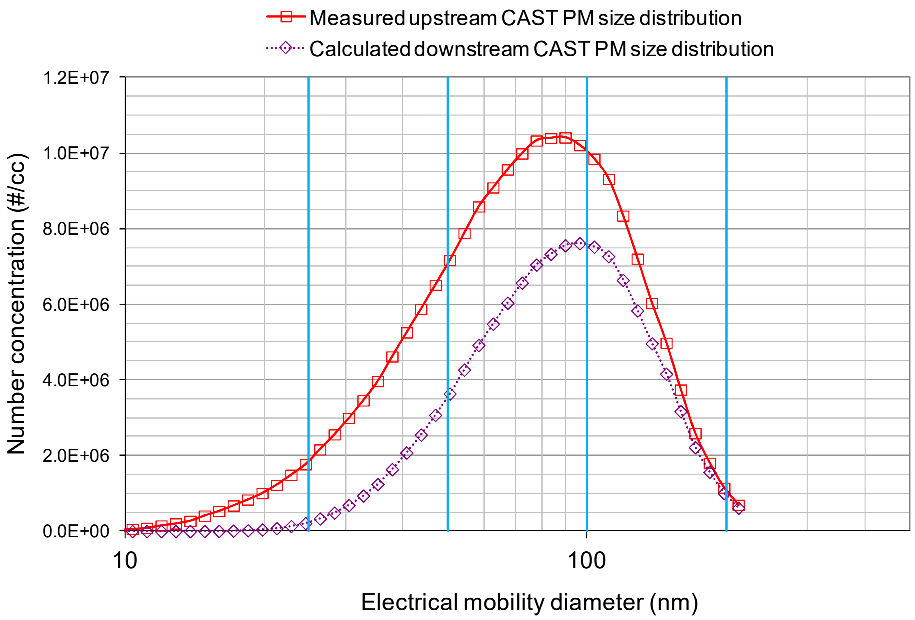
Schematic of set-up examining size-selected particle deposition efficiency

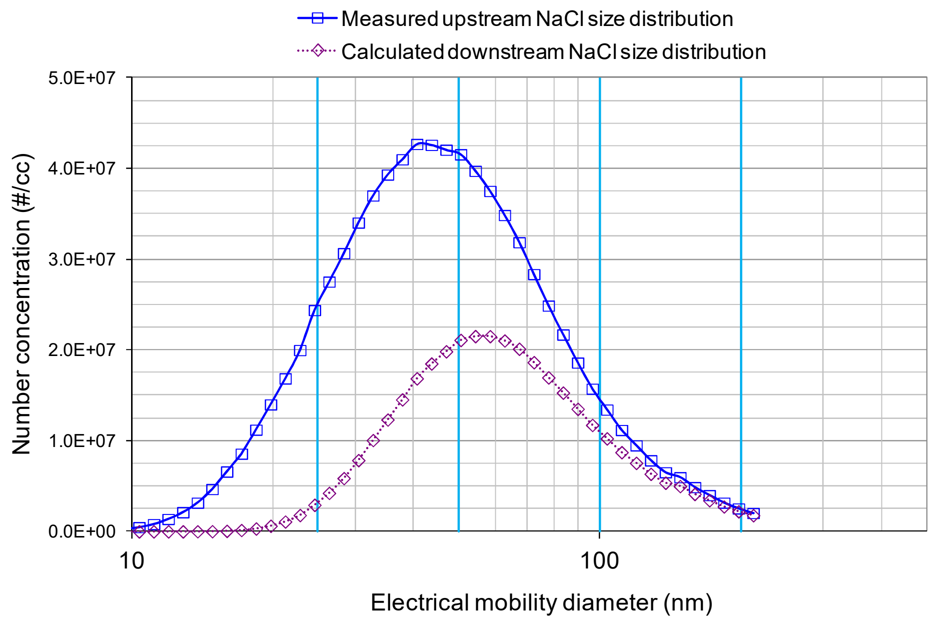


Measurement of penetration of size-selected particles

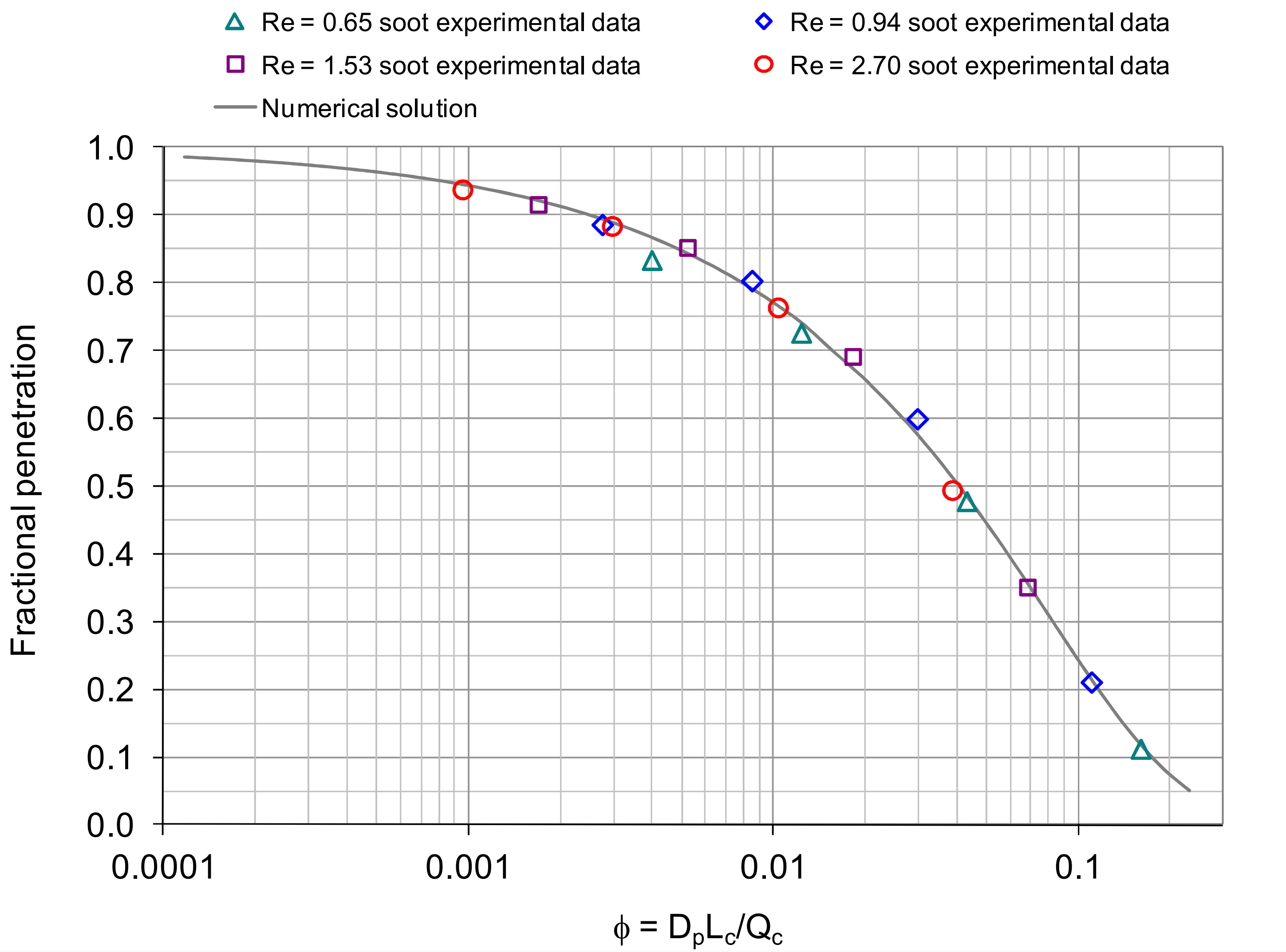
- Papaioannou et al. (SAE 2006-01-1075) previously used cordierite honeycomb monolith as diffusion battery to remove smallest particles from gas stream as part of selective particle sampler system
- In this study particles size-selected by DMA (25, 50, 100 and 200 nm) were sent through 600 cpsi, ϕ4.66” by 6” monolith at various flow rates (0.65 < *Re* < 2.70) relevant to typical mass transfer conditions in DPF porous wall
- Two aerosol sources used to investigate effect of particle morphology and interception length:

▼ SMPS size distributions of fractal-like CAST aggregates and cubic NaCl crystals (dispersed from aqueous solution and dried with dessicating column) plotted with calculated penetration through flow-through monolith after diffusion losses for *U* = 1 cm/s (*Re* = 0.65)





Efficiency of deposition of size-classified CAST aggregates (plotted with numerical solution for particle diffusion in square channels) vs deposition parameter ϕ



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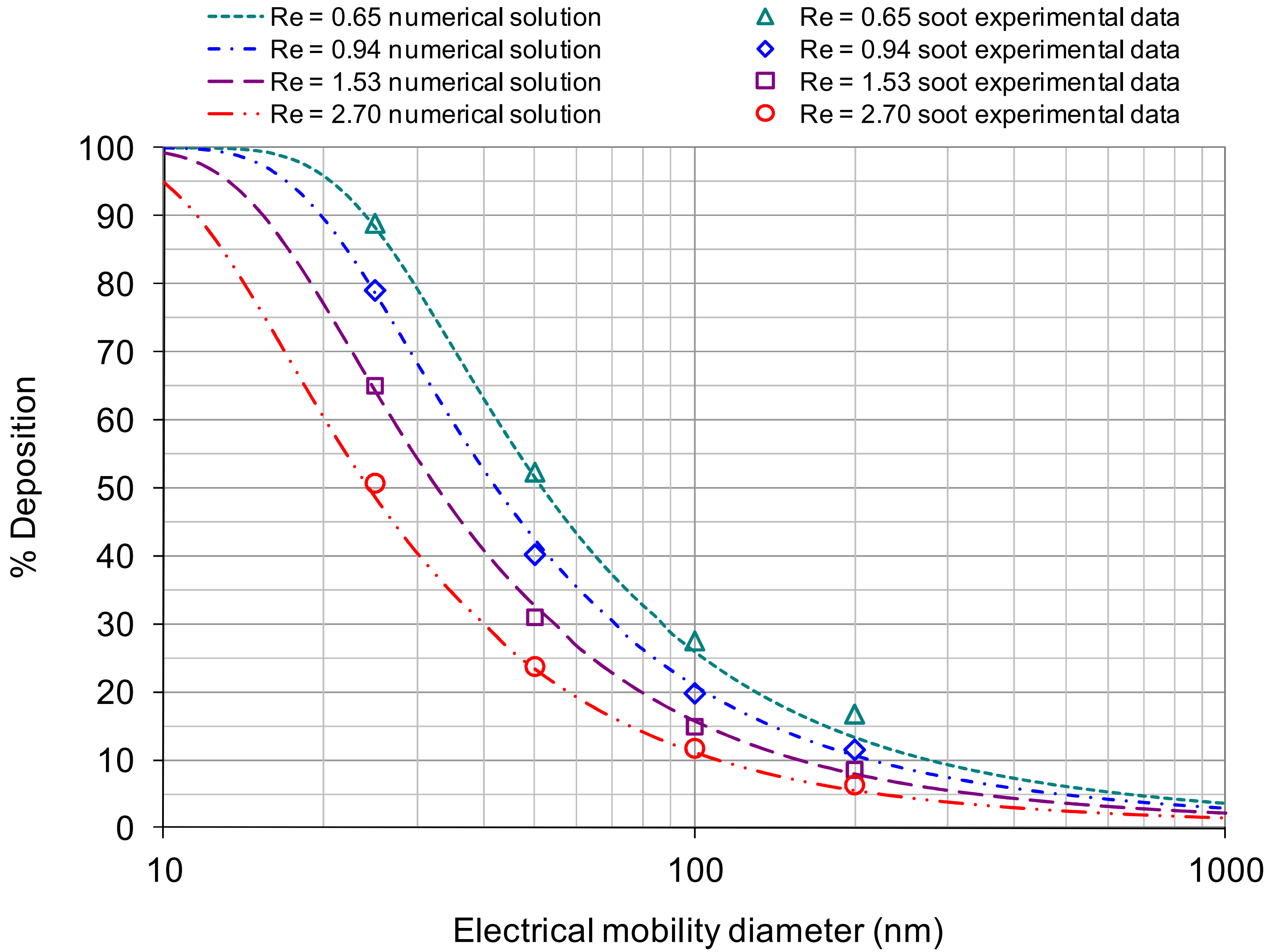
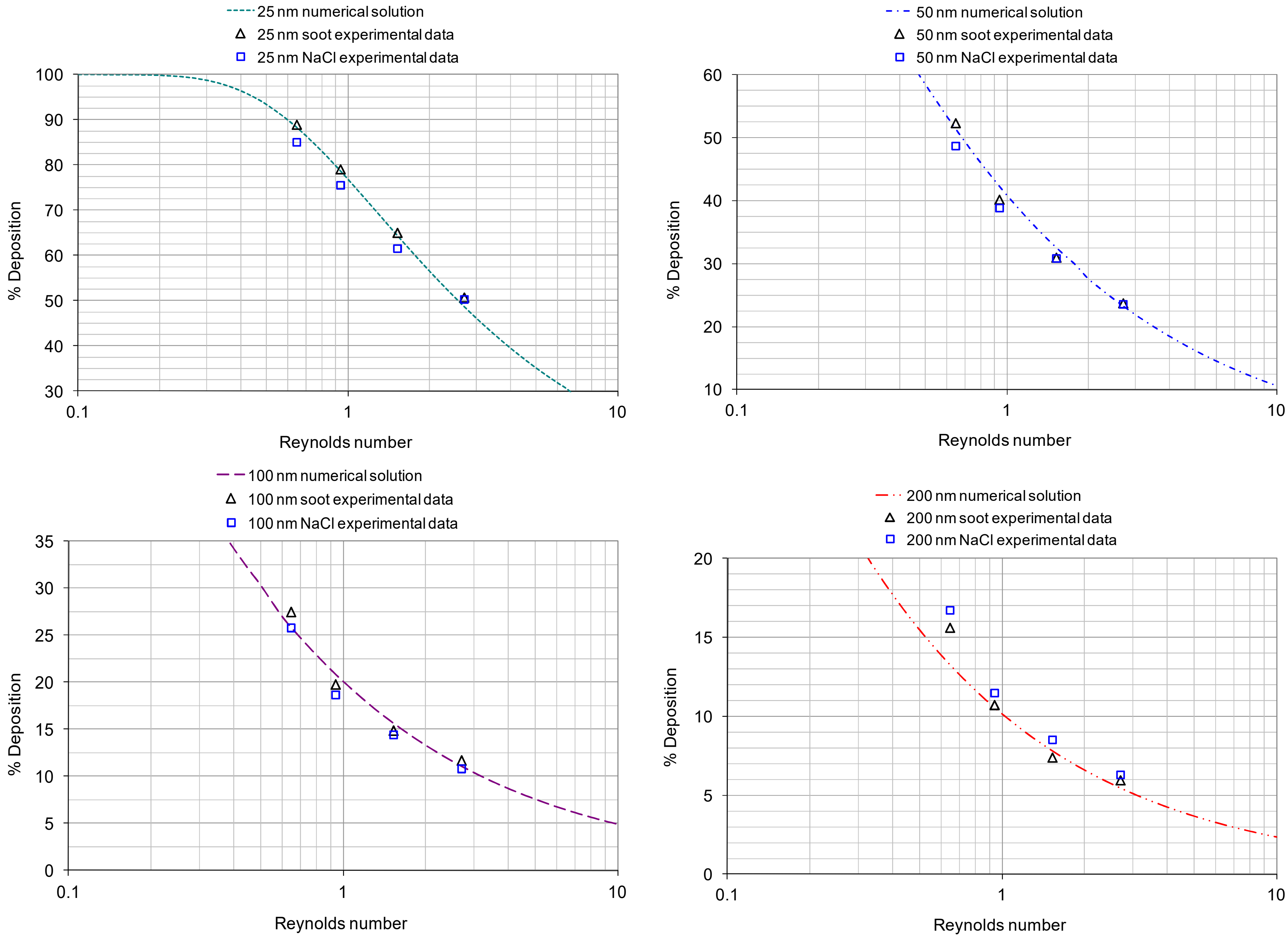
Efficiency of deposition of size-classified CAST aggregates (plotted with numerical solution for particle diffusion in square channels) vs electrical mobility diameter



Comparison of deposition efficiency for fractal-like CAST aggregates with cubic NaCl particles



Gravitational settling

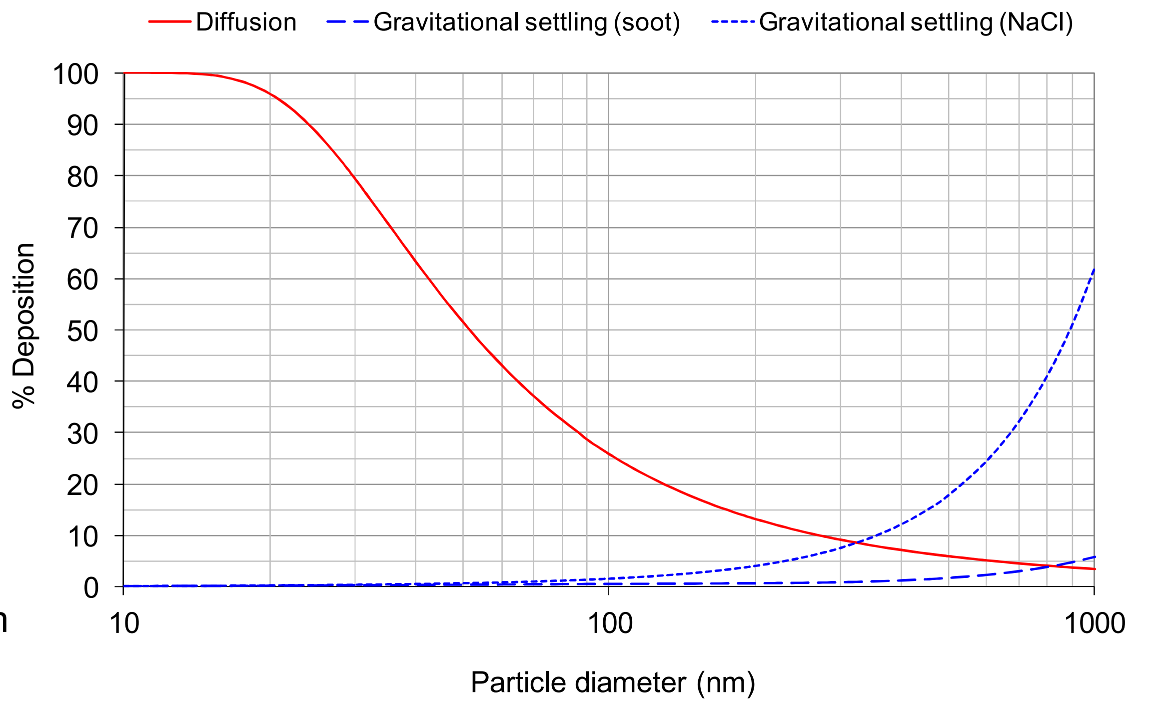
- Monolith channels are horizontally-oriented
- Most significant discrepancy between data and theoretical curves for diffusion occurred for largest selected mobility diameter (200 nm) and greatest residence time in channel
- At 200 nm, deposition significantly greater for denser NaCl crystals vs CAST aggregates

Critical trajectory of particle in horizontal, fully-developed laminar flow through single square channel leads to following expression for settling efficiency, E_G:

$$E_G = \frac{V_G \tau_C}{2d_h}$$

V_G – terminal settling velocity (balance of gravitational force with Stokes drag)

τ_C – residence time (= L/U)
d_h – hydraulic diameter



This calculation leads to combined deposition efficiency (assuming diffusion and settling act independently) of 16.8% for 200 nm NaCl at U = 1 cm/s (STP) where experimental value was 16.7%

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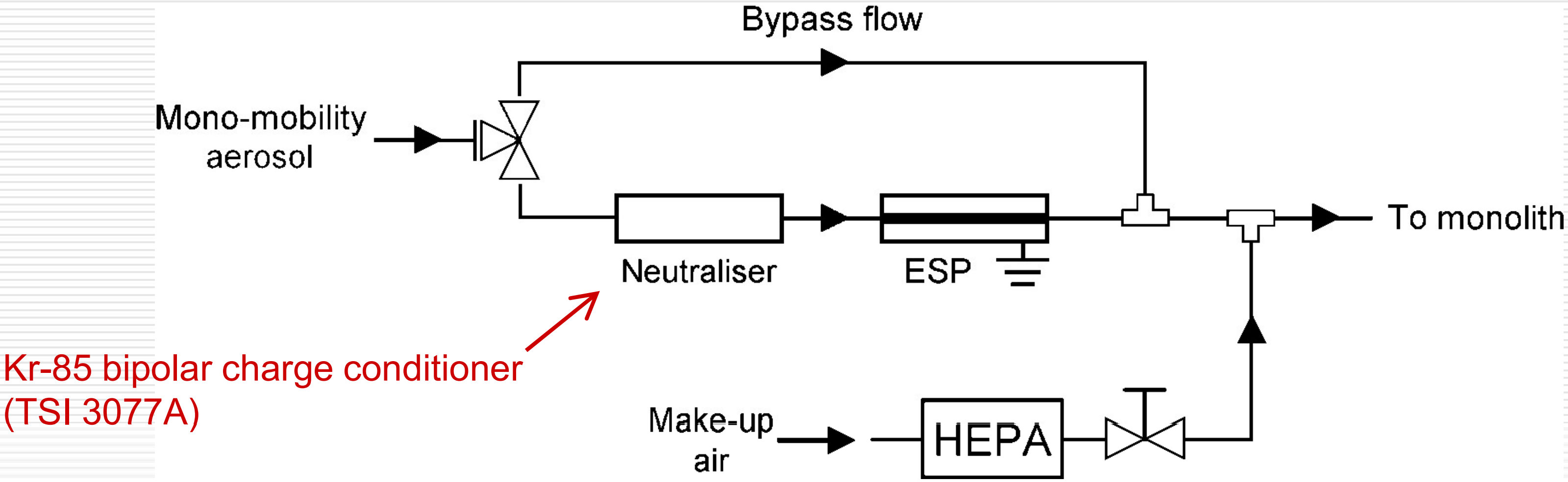
Effect of aerosol charge state on deposition in monolith

Schematic of modified set-up:



Boltzmann charge distribution prescribed by neutraliser:

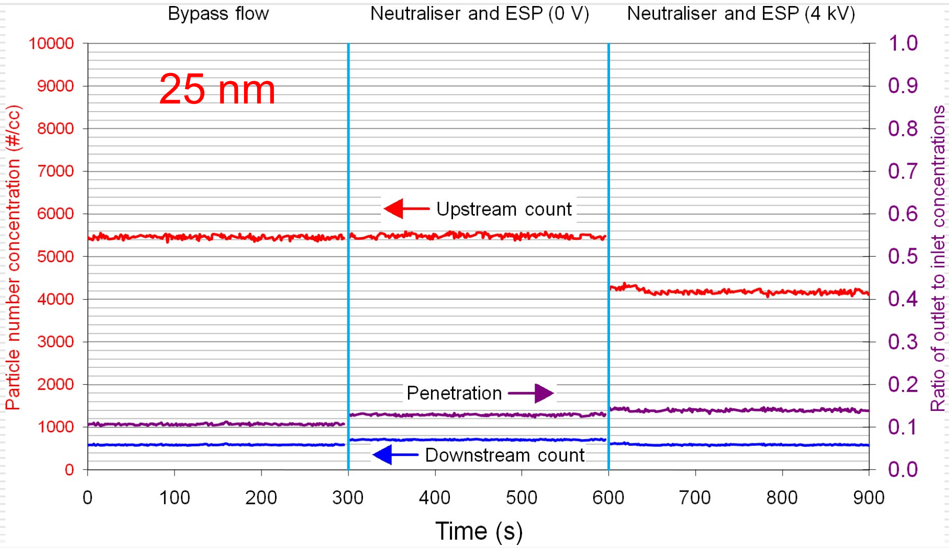
Mobility midpoint ($m^2V^{-1}s^{-1}$)	d_m for n_e = 1 (nm)	% total particle concentration carrying the following number of charges:								
		-4	-3	-2	-1	0	+1	+2	+3	+4
3.678×10^{-3}	25	0.00	0.00	0.07	13.23	76.51	10.16	0.03	0.00	0.00
9.840×10^{-4}	50	0.00	0.00	1.08	21.96	59.64	16.71	0.62	0.00	0.00
2.823×10^{-4}	100	0.00	0.33	5.35	27.79	42.09	21.27	3.02	0.15	0.00

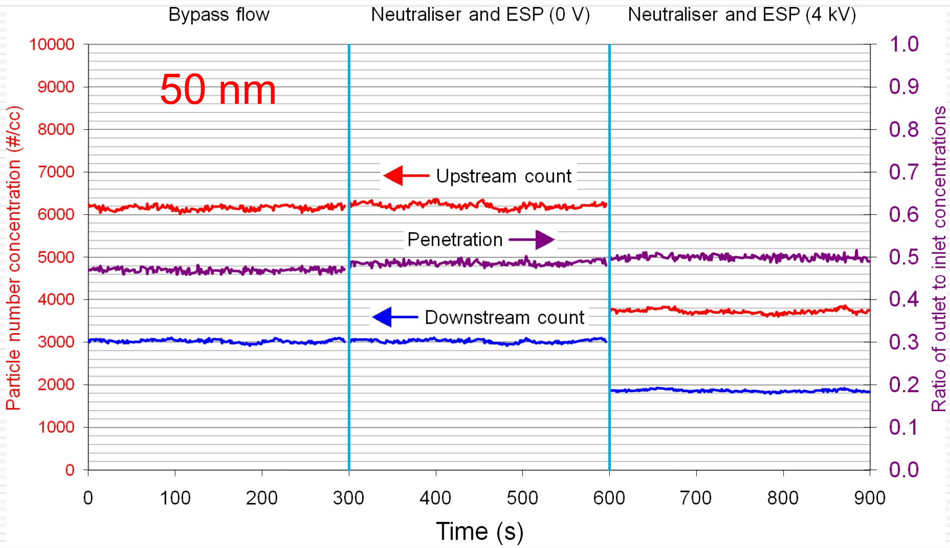
Deposition efficiencies at *U* = 1 cm/s (*Re* = 0.65)

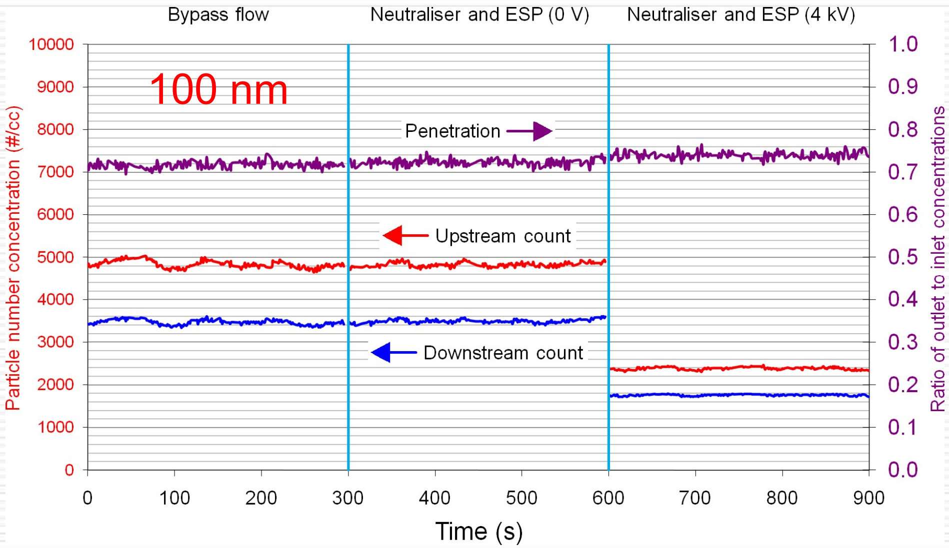
1. Bypass flow: 100% positively charged particles from DMA (mostly singly-charged)

2. Neutraliser and ESP (0V): Kr-85 charge-conditioned particles (**76%** uncharged for 25 nm, **60%** uncharged for 50 nm, **41%** uncharged for 100 nm)

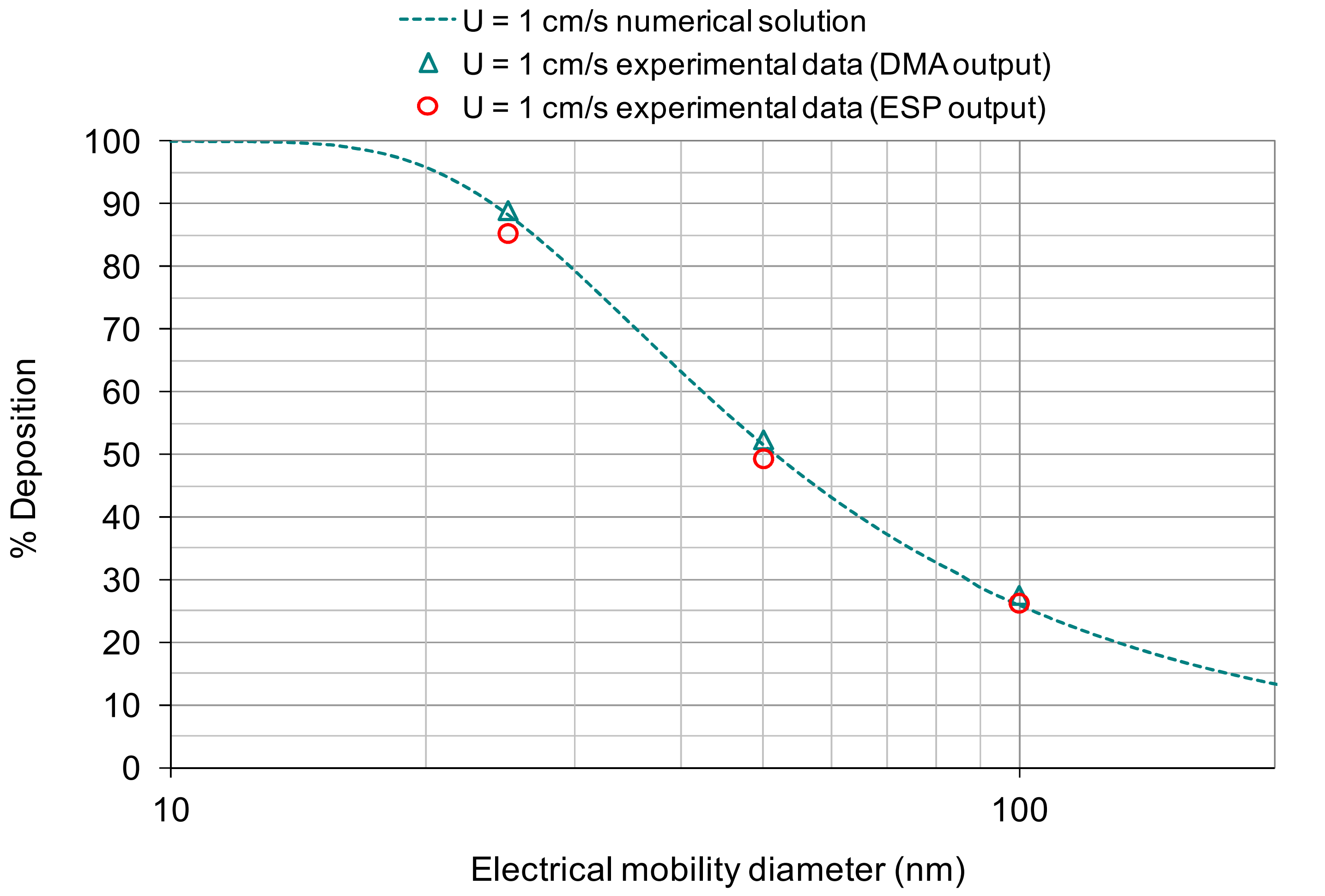
3. Neutraliser and ESP (4 kV): **100%** uncharged







Deposition efficiencies at *U* = 1 cm/s (*Re* = 0.65)



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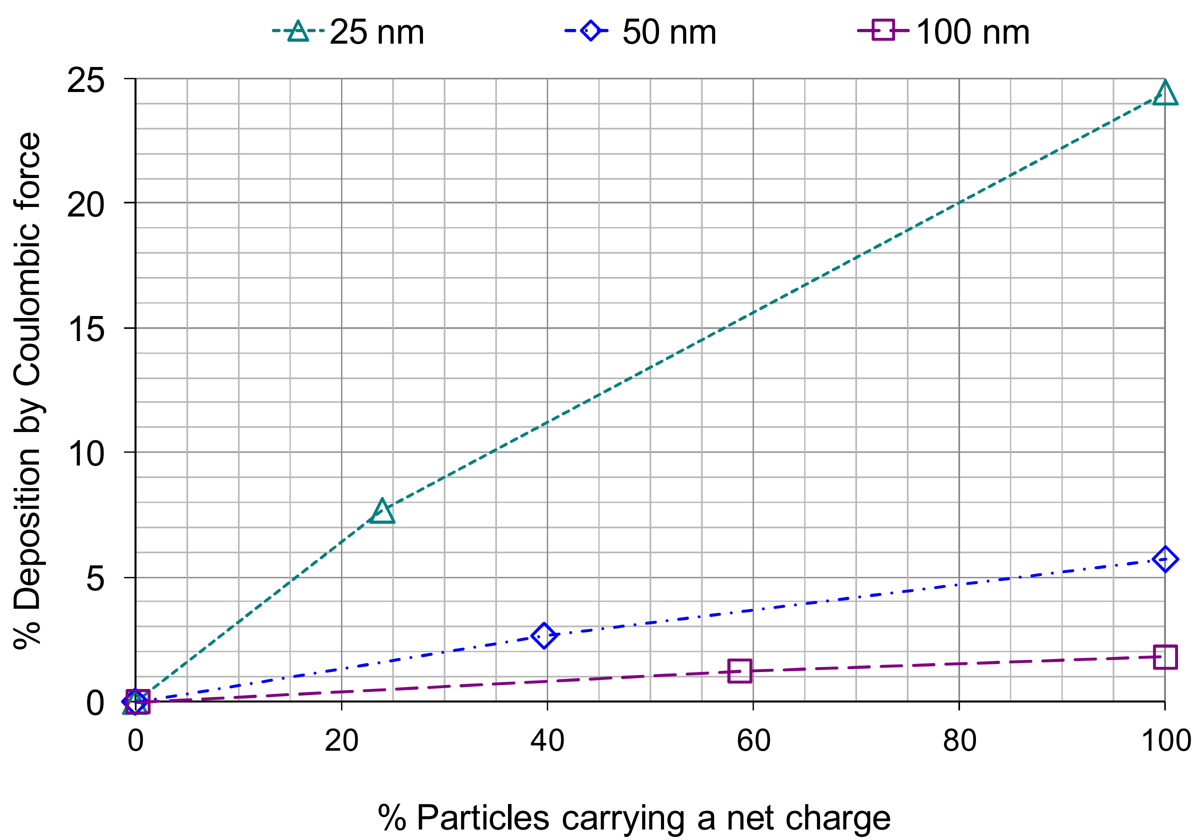
Deposition by Coulombic force

Taking the aerosol emerging from the ESP as the baseline case, the below graph shows the fraction of uncharged particles penetrating the monolith that then precipitate when the aerosol acquires charge:



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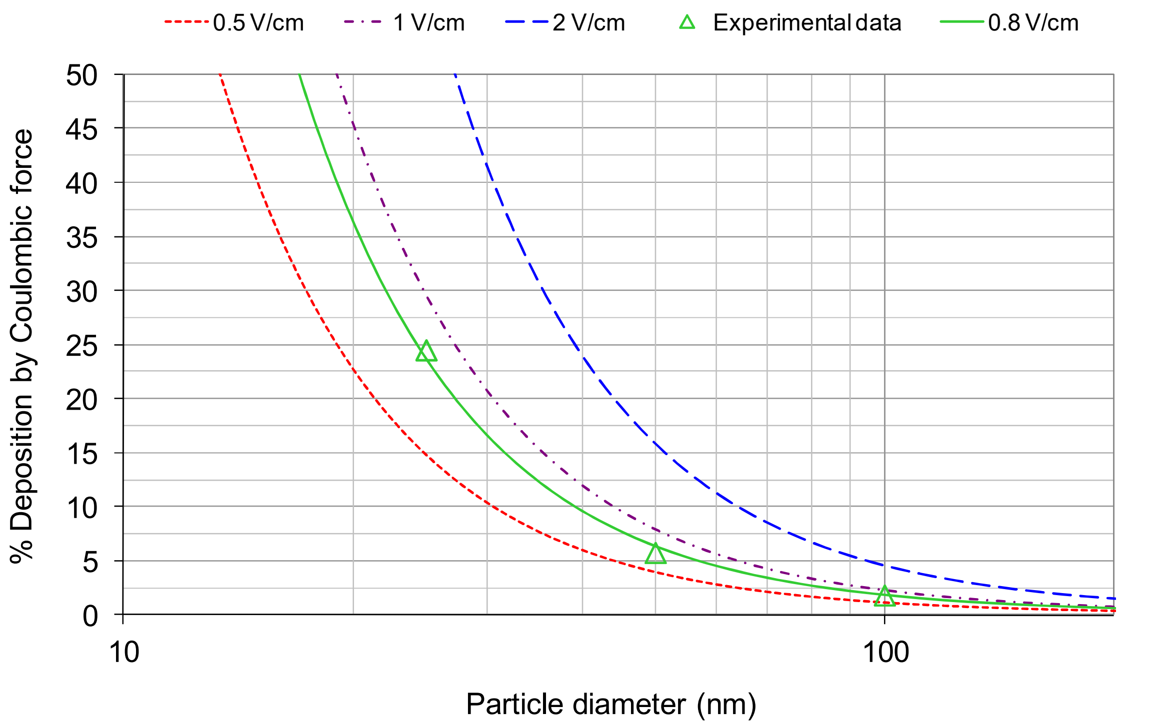
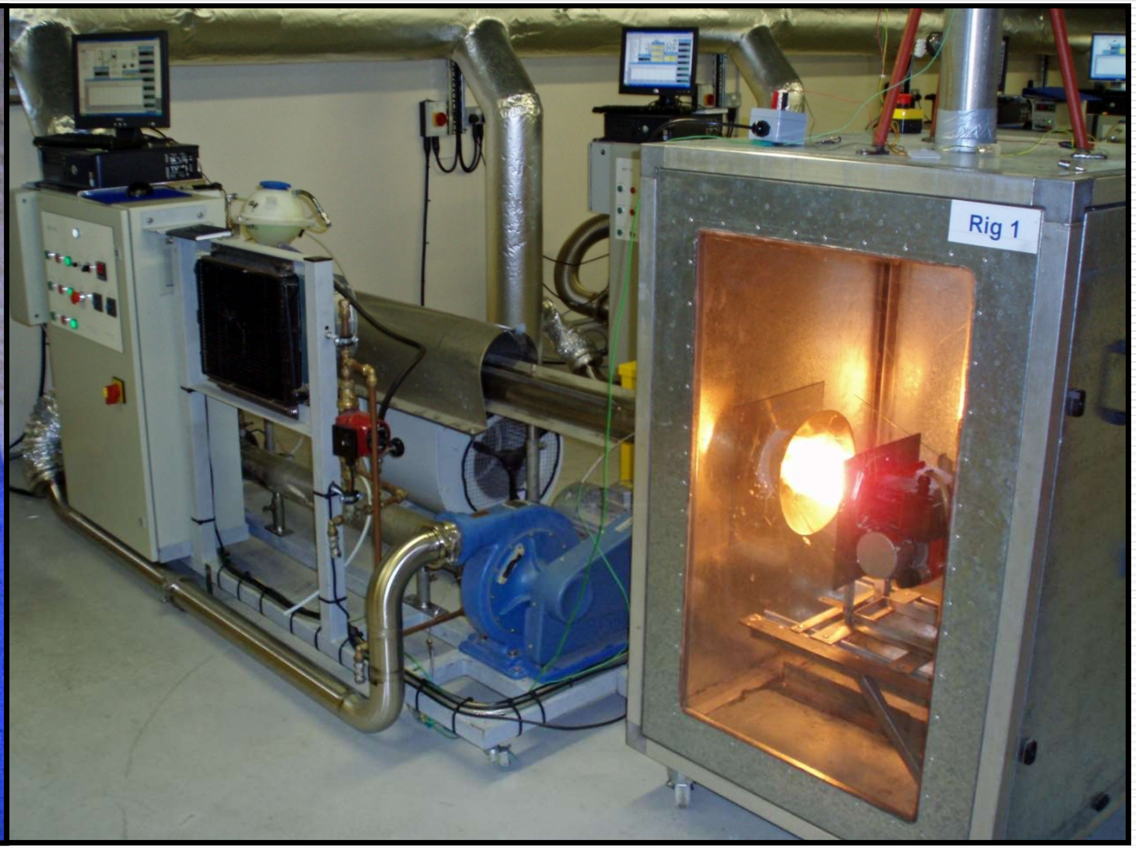
Deposition by Coulombic force

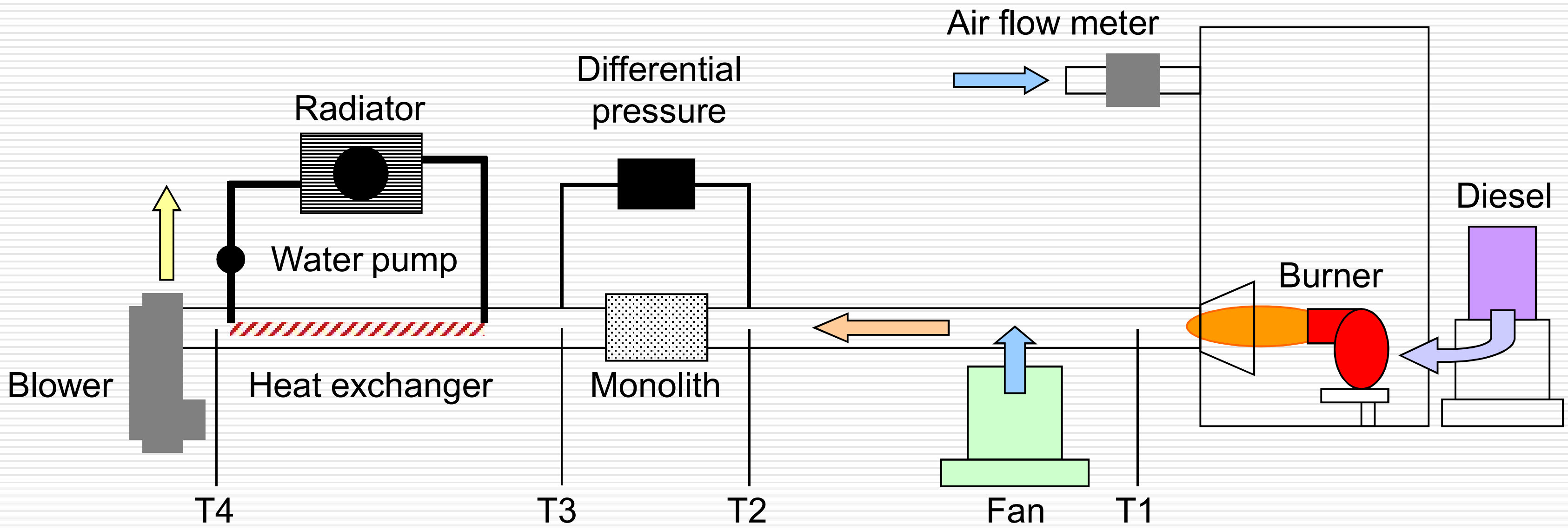
- Precipitation of singly charged particles is due to weak electric fields arising from static charges retained on non-conductive cordierite surface
- Their action can be calculated using same expression for gravitational settling but replacing V_G with V_E (= electrical mobility x field strength) assuming fields act across entire width of channel i.e. not affected by adjacent charges (Applied Catalysis B: Environmental 10 (1996) 117-137)
- Calculations for several field strengths plotted below along with experimental values for unconditioned DMA output:



Loading of monoliths at light-duty engine exhaust flow rates with Johnson Matthey diesel burner



Johnson Matthey diesel burner

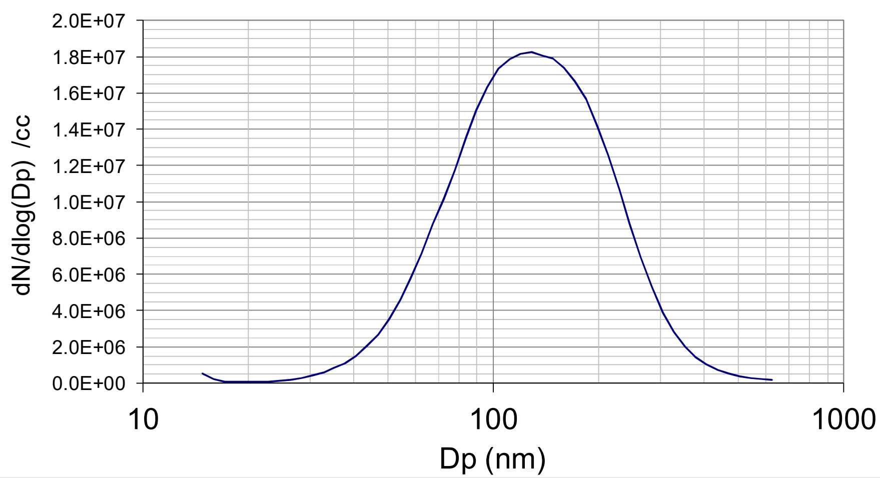


Exhaust volume flow rate: 2.35 m³/min
(correct at 0°C and 1013.25 mb)

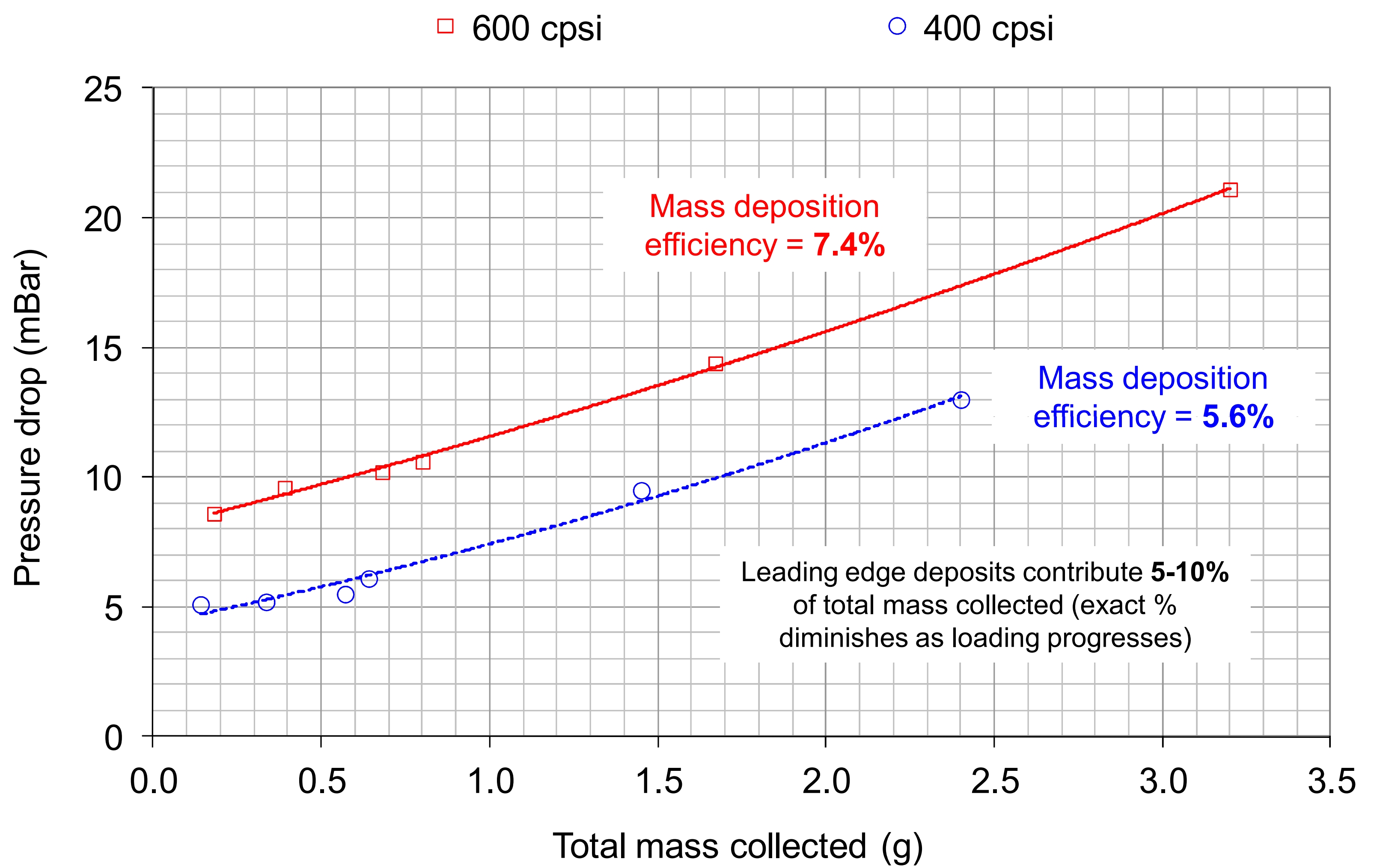
Particulate loading rate: ~5 g/hr
(exact value monitored with TEOM)

Monolith inlet temperature: 220-250°C

Size distribution ►
(TGA analysis: 73.5 wt% EC, 27.4 wt% OC)

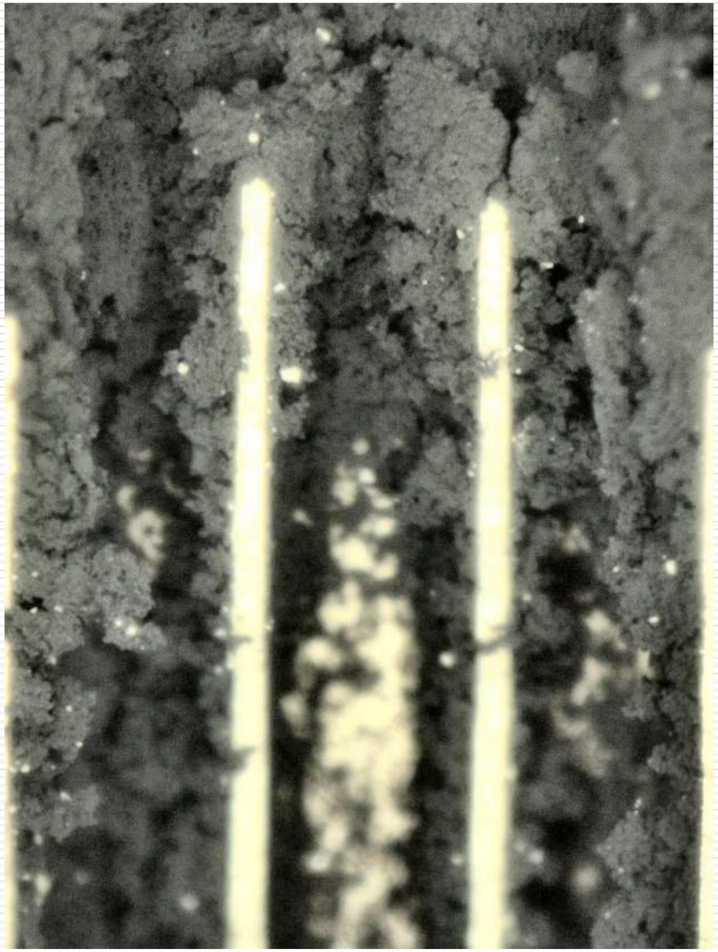


Pressure drop v diesel particle mass collected for flow-through monoliths (ϕ5.66 in by 6 in) loaded continuously for up to 10 hours



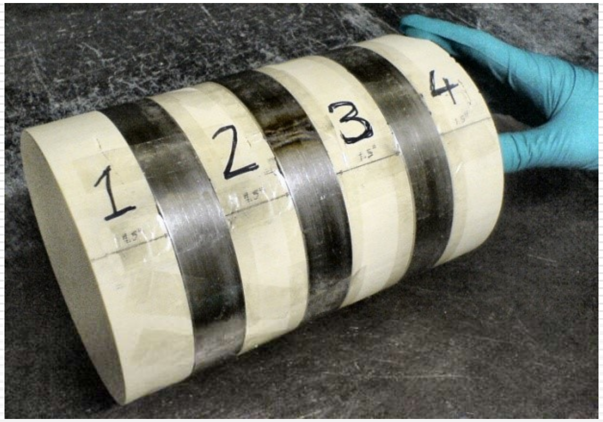
Optical micrographs of leading edge deposits on 600 cpsi monolith

- 6 hours' loading: 29.6g incident particulate mass and 1.7g collected
- Distance between centre points of adjacent cells is 1.04 mm

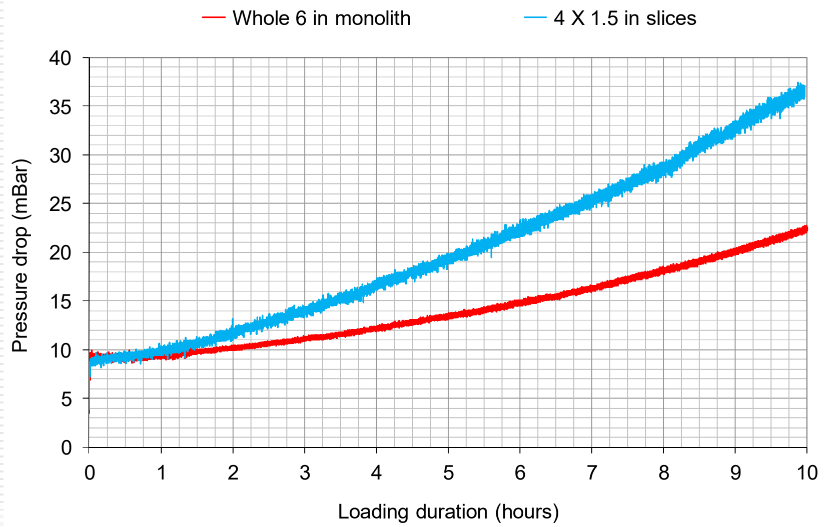


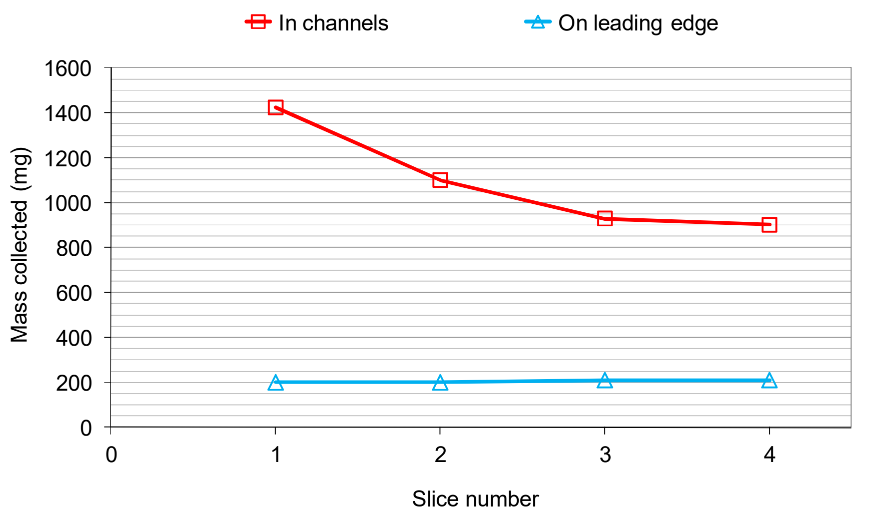
ΔP and diesel particle mass collected in four 600 cpsi monolith slices

- Monolith radially sawn into 4 x 1.5 in long slices and inserted into exhaust can alternately with 3 x 1 in long steel ring spacers:

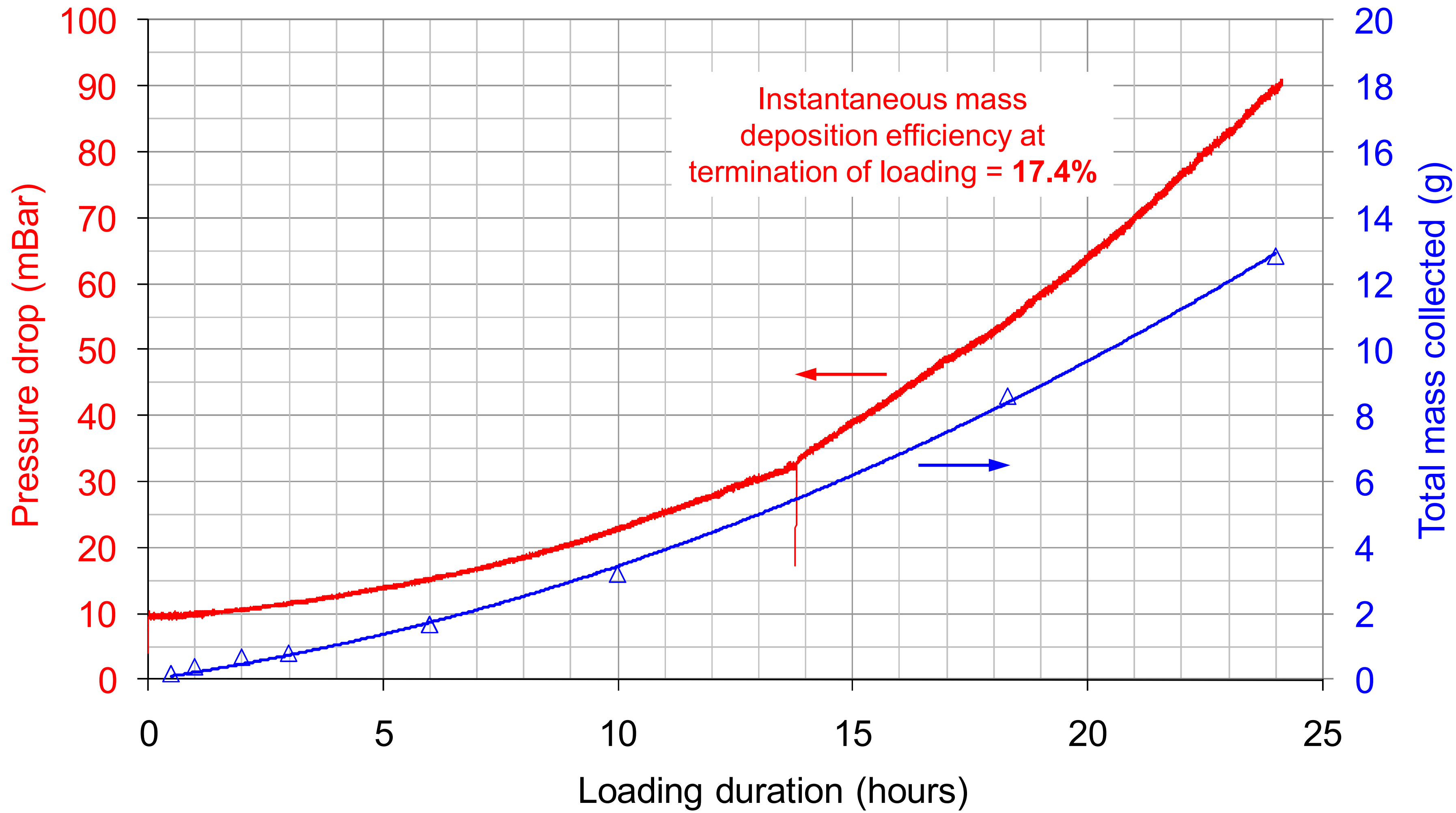


- Re-distribution of flow between slices and addition of 3 leading edges significantly increased overall mass collection vs whole monolith
- Leading edge deposits are constant by slice
- 1st slice represents 1st quarter of whole monolith, where 44% of total mass collected is found

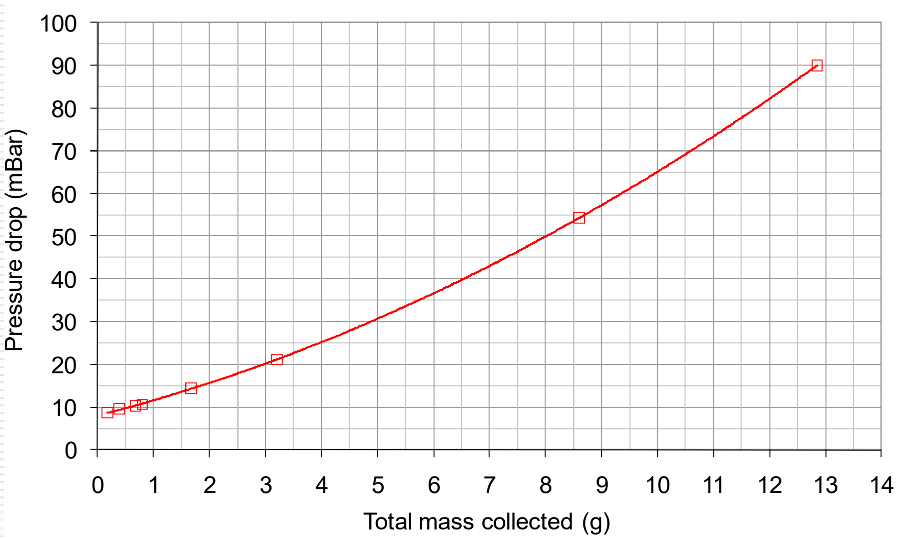


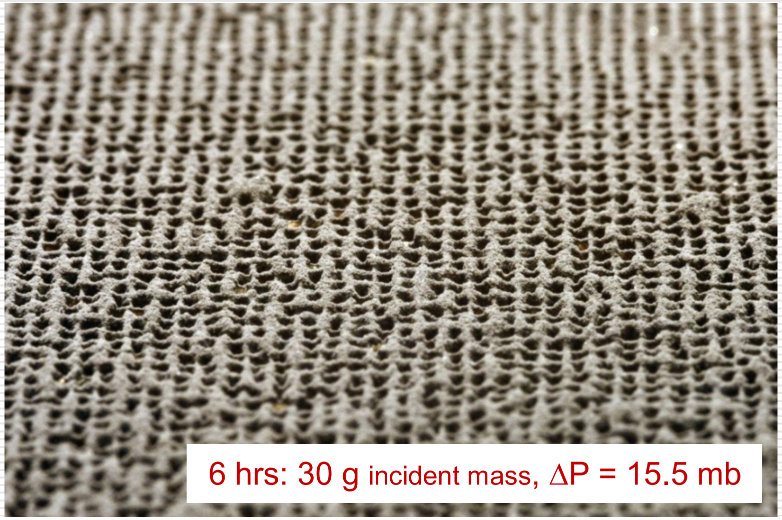


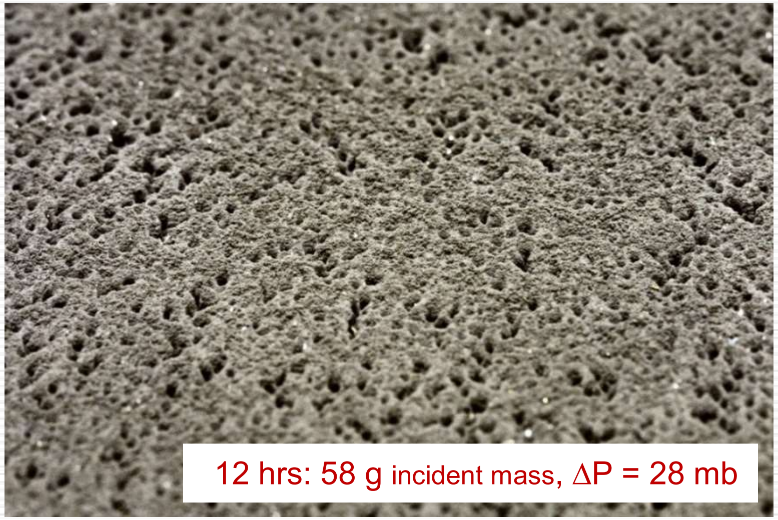
Successive loading of 600 cpsi monolith up to max ΔP (limited by blower)

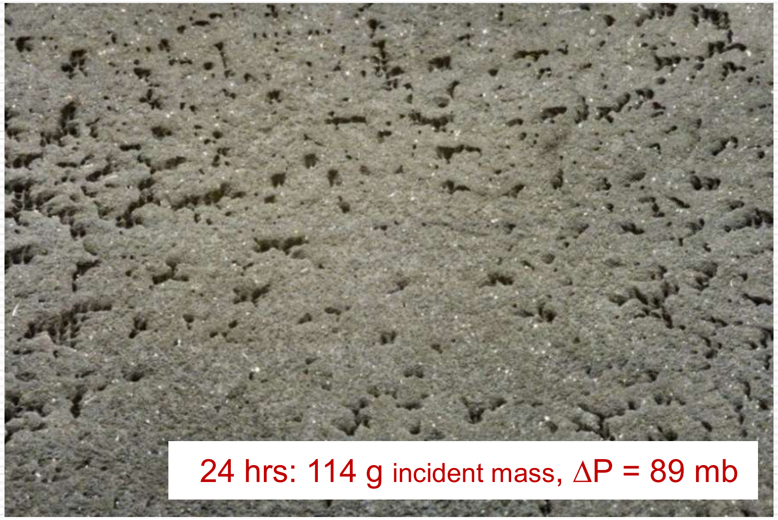


ΔP v diesel particle mass collected for 600 cpsi monolith loaded for 24 hours









Conclusions

- Deposition of singly charged CAST aggregates for Reynolds numbers below 5 and flow velocities below 10 cm/s (in the range of mass transfer conditions occurring in the pores of a wall-flow DPF) showed close agreement with numerical solutions for diffusion of uncharged spheres from fully developed laminar flow to the walls of a square channel.
- CAST aggregate deposition was slightly greater than for cubic NaCl crystals for 25-100 nm due to greater interception length, but at 200 nm gravitational settling of higher density NaCl became appreciable.
- Deposition of DMA output aerosol was slightly reduced when it acquired a Boltzmann charge distribution and reduced again when it passed through an ESP; when the action of the Coulombic force was isolated from diffusion and quantified, good agreement was found with calculated particle capture due to weak electric fields arising from static charges on the cordierite surface.

- Long-term particulate loading of uncoated monoliths at typical light-duty diesel exhaust flow rates showed that instantaneous mass collection can eventually approach 20%; the flow through the monolith remains predominantly Darcian while few channel entrances are obstructed then pressure losses scaling with U^2 are incurred when significant portions of cake form.
- Update: further work to model diesel particle deposition in flow-through monoliths (interception at leading edge and coupled diffusion-interception inside channels) has been published – see Haralampous and Payne, Int. J. Eng. Res. 14 (2016) 1-17

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Thank you for your attention