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After-treatment of Nanoparticle Emissions from Gasoline Engine Exhausts

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High numbers of ultrafine particles in ambient air have been associated with respiratory effects and deaths [1,2]; biological research suggests that particle surface area and chemistry are more important factors than mass, and particles not toxic in micron sizes may be toxic as ultrafine (< 100 nm) and nanoparticles (< 50 nm) [3]. Hence PM emissions from gasoline as well as diesel engines are of growing concern, in view of evidence from laboratory and roadside measurements that gasoline vehicles emit a higher proportion of nanoparticles than diesel vehicles at high speeds and loads [4, 5] and in numbers which are comparable to those from diesel vehicles. Gasoline direct injection (DISI or GDI) engines may emit higher particle numbers than modern port fuel injected (PFI) engines [6, 7].

The objective of this project, undertaken jointly with Johnson Matthey, U.K., is to investigate the most effective techniques for nanoparticle capture in the exhaust and the most suitable catalysts for captured PM destruction, *i.e.* trap regeneration, in order to develop a device for gasoline PM after-treatment.

Diffusion and thermophoresis were found to be less effective than electrostatic fields as mechanisms for nanoparticle collection in gasoline engine exhaust systems [8, 9]. Filtration has been investigated, as it may be a reliable particle trapping process provided that cost-effective solutions can be found for the filter regeneration and increased engine back pressure. Filters are very effective at collecting PM by mass [10] but they may increase PM by number under certain conditions [11].

This presentation reports an evaluation of the performance of a ceramic wall-flow filter (Corning EX-80-200-12) and a prototype wire-cylinder electrostatic precipitator in reducing particle concentrations by number. PM was simulated by synthetic carbon particles generated by spark discharge from a Palas GFG1000 carbon aerosol generator, in air flow at temperatures from 20°C to 400°C. The wall-flow filter has also been investigated with engine-out PM from a Mitsubishi Carisma 1.8 litre GDI-engined passenger car on a chassis dynamometer. A TSI 3969 Scanning Mobility Particle Sizer was used for characterizing particles in the PM simulation while a Dekati Electrical Low Pressure Impactor was used for the GDI engine tests, samples being diluted using a Dekati two-stage ejector dilutor [12].

Collection efficiencies by number from 65 % to 85 % have been obtained from the wall-flow filter with simulated PM, for long filter exposure time (~ 20 h) at 20°C and 400°C with particle mass loading typical of gasoline engine exhausts. Effective particle number reductions were obtained with the same type of filter in the exhaust of the GDI-engined car for both steady state operating conditions and during transients. A result for the ECE+EUDC drive cycle is shown in Figure 1. Only during the high-speed part of the EUDC cycle (120 km/h) were post-filter particle numbers similar to those in the baseline case without filter [12].

Suitable catalysts have been identified by the industrial partner for the destruction of captured PM under typical low-oxygen gasoline-engine exhaust conditions [12]. Although the wall-flow filter showed promising results for capture of both simulated PM and GDI engine-out PM, the collecting electrode wall of an ESP is likely to provide a better support for such catalysts, which have a mobile active phase.

A prototype wire-cylinder electrostatic precipitator (ESP) has been constructed, consisting of a grounded stainless-steel cylinder of 50 mm diameter and 1 m length as the collecting electrode with an axial discharge electrode of 0.1 mm diameter tungsten wire. The dimensions were selected on the basis of estimates of the axial distance required for 100% particle capture at typical gas velocities in the laboratory PM simulation, supported by analytical modelling in progress [9]. The wire is kept both taut and centrally located by an insulated tensioning device, which allows the ESP to be installed in line with the simulated exhaust pipe, in place of the wall flow filter. Particles entering the ESP are first charged by corona discharge and then transported by electrophoretic drift to the pipe walls, where deposition occurs.

Results are presented for the uncoated ESP and a catalyst-coated ESP at a fixed simulated carbon particle load, for different gas flow conditions, residence time, temperature up to 400°C and electrode voltage up to +14 kV. No negative effect of the catalyst coating on ESP operation has been observed. Little variation occurred in particle concentration over the pipe cross section at ESP inlet or outlet, and particle capture was confirmed by observation of uniform black deposits. Collection efficiency by number exceeded 95% when operating the catalyst-coated ESP at an electrode voltage of +8 kV at gas temperature up to 400 °C and short gas residence times (<1 s), as illustrated in Figure 2. Electrical power consumption was negligible. Engine testing of the catalysed ESP is in progress.

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References

1. Seaton, A., MacNee, W., Donaldson, K. and Godden, D., , *The Lancet*, Vol. 345, pp 176–178 (1995)
2. Wichmann, H. E., Spix, C. *et al*., Rep. 98, Health Effects Inst., Cambridge, Mass. (2000)
3. Osunsanya, T., Prescott, G. and Seaton, A., *Occup. Environ. Med*., Vol. 58, pp154-159 (2001)
4. Hall, D. E., Goodfellow, C. L., Heinze, P. *et al*., SAE Paper 982600 (1998)
5. Kayes, D. and Hochgreb, S., , *Environ. Sci. & Tech*., Vol. 33, pp 3957-3967 (1999)
6. Graskow, B. R., Kittelson, D. B. *et al.*, SAE Paper 1999-01-1145 (1999)
7. Graskow, B. R., Kittelson, D. B. *et al.*, SAE Paper 1999-01-1144 (1999)
8. Bouris, D., Arcoumanis, C. and Bergeles, G., Instn Mech. Engrs Conf. "21st Century Emissions Technology", London, 4-5 Dec. (2000)
9. Rubino, L., Shrimpton, J. S. and Crane, R. I., 17th Ann. Conf. on Liquid Atomization & Spray Systems, Zurich, 2-6 Sep. (2001)
10. Hawker, P., Huthwohl, G., Henn, J. *et al.*, SAE Paper 980189 (1998)
11. Kruger, M., Luders, H., Luers, B. *et al*., *Motortechnische Zeitschrift*, Vol. 58 (1997)
12. R. I. Crane, L. Rubino, C. Arcoumanis, S. E. Golunski *et al*., "Strategies for Gasoline Particulate Emission Control – a 'Foresight Vehicle' Project", SAE Paper 2002-01-1894 (2002)

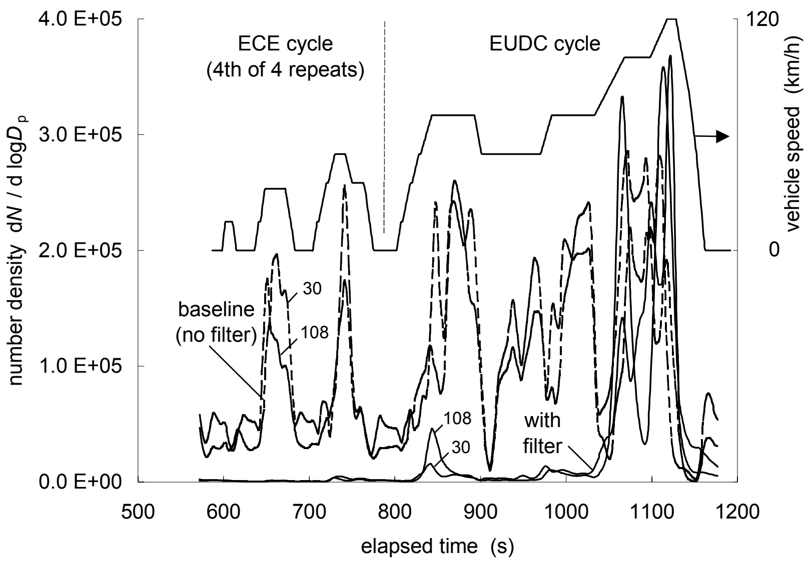

Figure 1

Measurements of wall-flow filter performance on GDI-engined vehicle over European ECE + EUDC drive cycle : time variation of particle number density for selected ELPI size bands, sizes (in nm) denoted by numbers beside curves

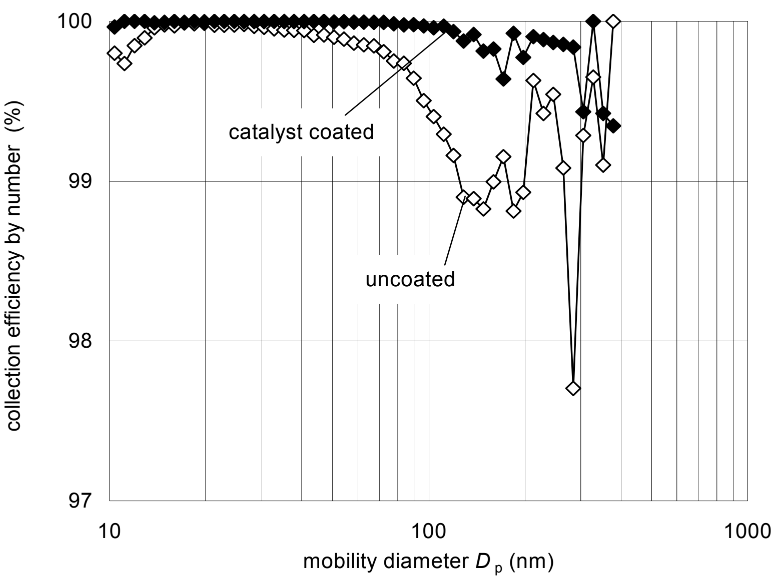

Figure 2

Collection efficiency by particle number of uncoated and catalyst coated ESP prototype;

ESP inlet temperature 400°C, central wire electrode at 8 kV, residence time 0.64 s

N.B. False origin on vertical axis

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Outline

- Objective
- Particle capture mechanisms & possible techniques
- Experimental investigation on effects of:
  - ◁ Wall-flow Filters
  - ◁ Prototype ESP
  - ◁ Integrated Catalysts / ESP

  on:
  - Simulation of gasoline engine PM
  - PM from GDI-engined vehicle
- Conclusions & Future Work

Objective

- Investigation of techniques capable of reducing gasoline PM number emissions & development of an after-treatment device for gasoline engined vehicles applications

Imperial College

Experimental & numerical investigation of gasoline PM capture and deposition in the exhausts

Johnson Matthey

Investigation of gasoline PM destruction using a catalyst under gasoline exhaust conditions

(SAE 2002-01-1894)

Particle Capture Mechanisms & Possible Techniques

- Gravitational settling
- Inertial impaction & interception
- Diffusion
- Thermophoresis
- Electrostatic fields
  - make it possible to alter particle trajectories

Ceramic filters

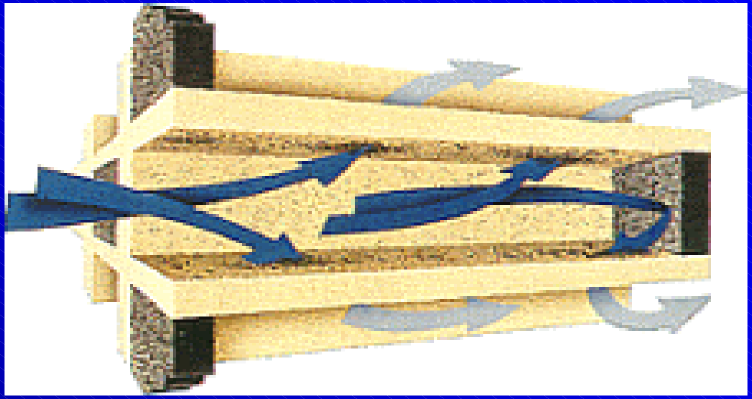
Cooler ?? …but Thermophoresis plays an important role only in rather slow flows with strong grad T

Electrostatic precipitators (ESPs)

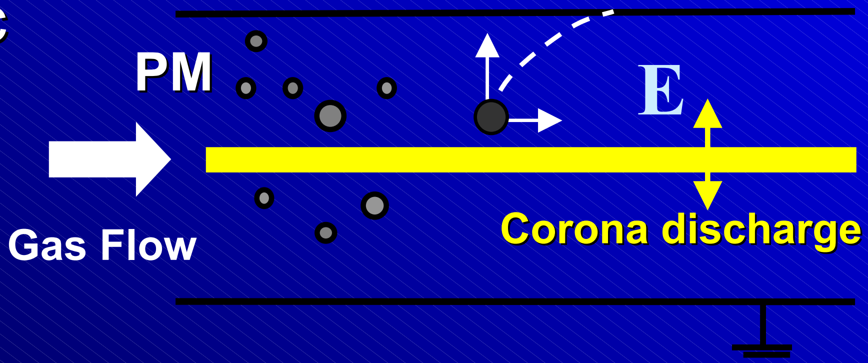
Experimental Investigation

- Simulation of gasoline engine particulates
  (control of variables can be difficult when source of PM is an engine)
  - Effects on particle number concentration and size distribution of:

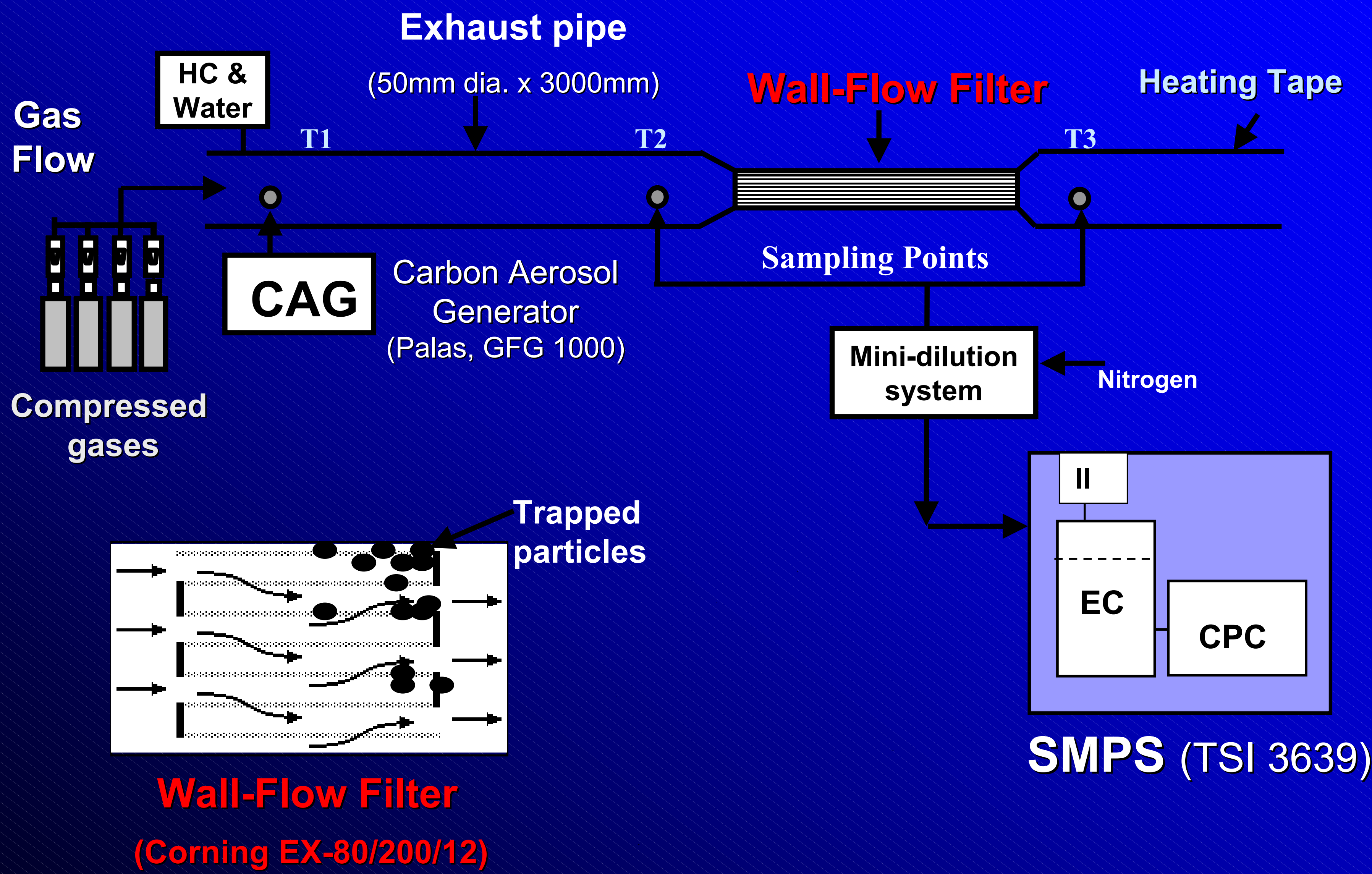
◁ Cordierite Wall-Flow Filter



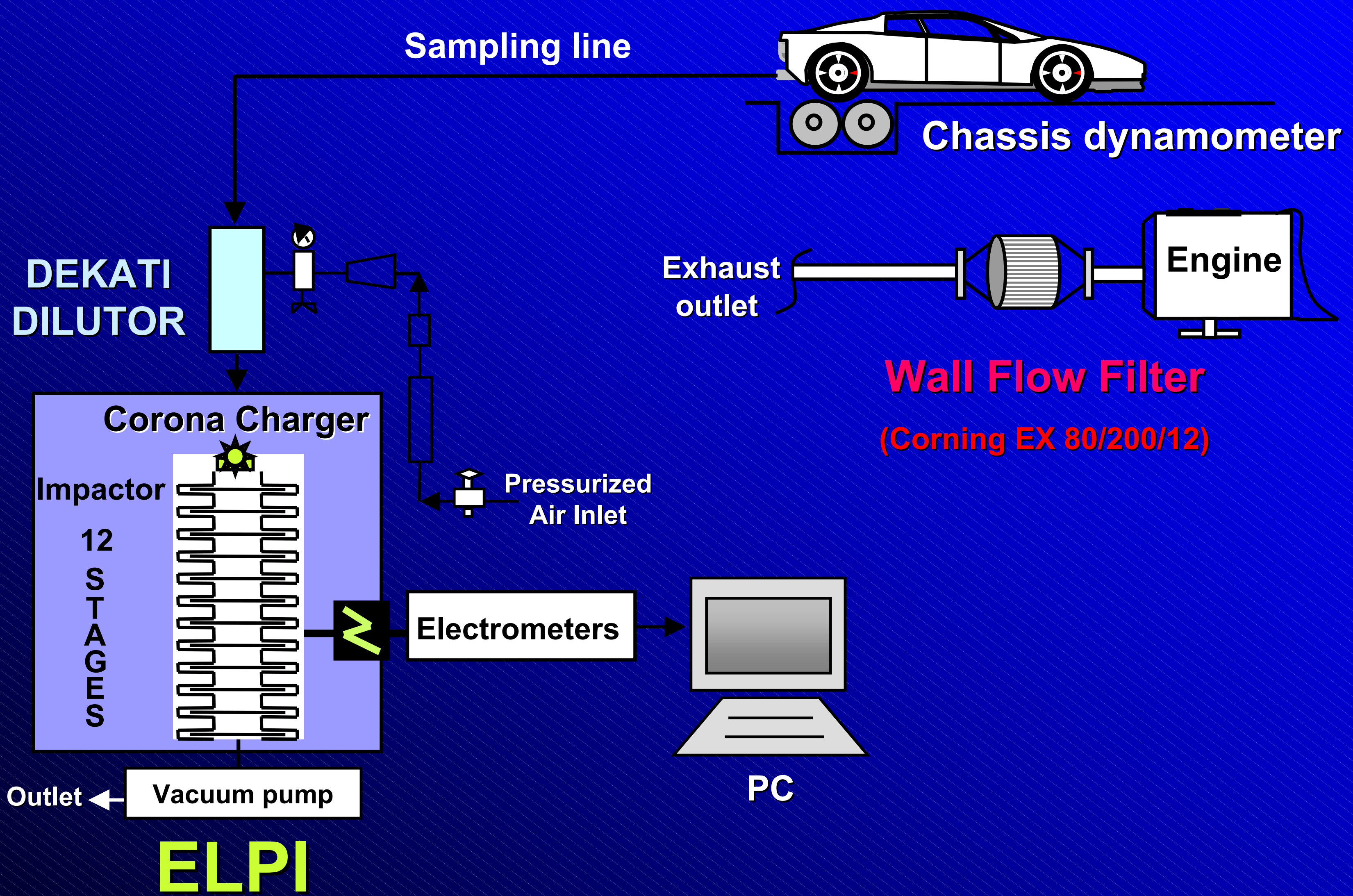
◁ Wire-Cylinder Electrostatic Precipitator (ESP)



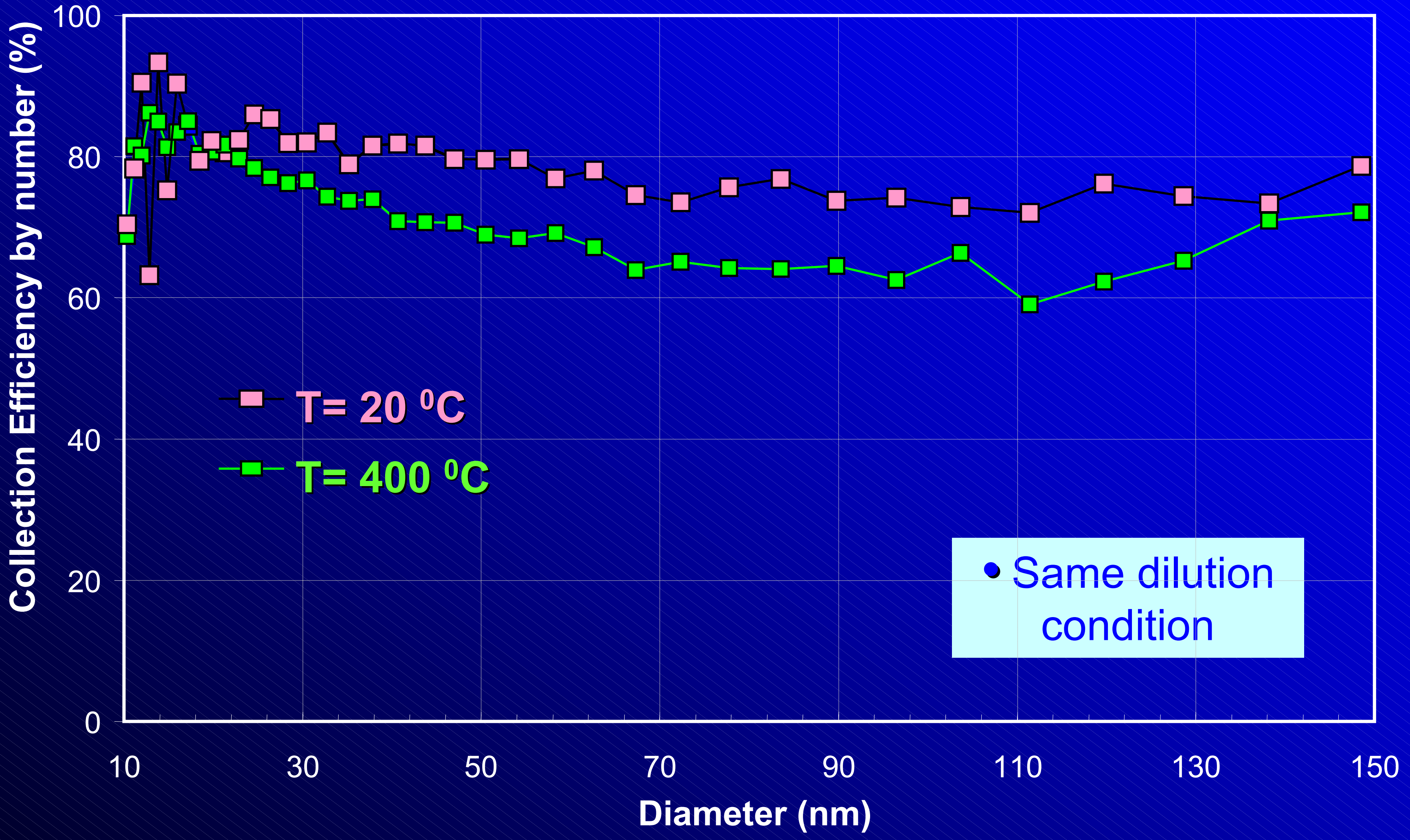
Simulation of gasoline engine PM



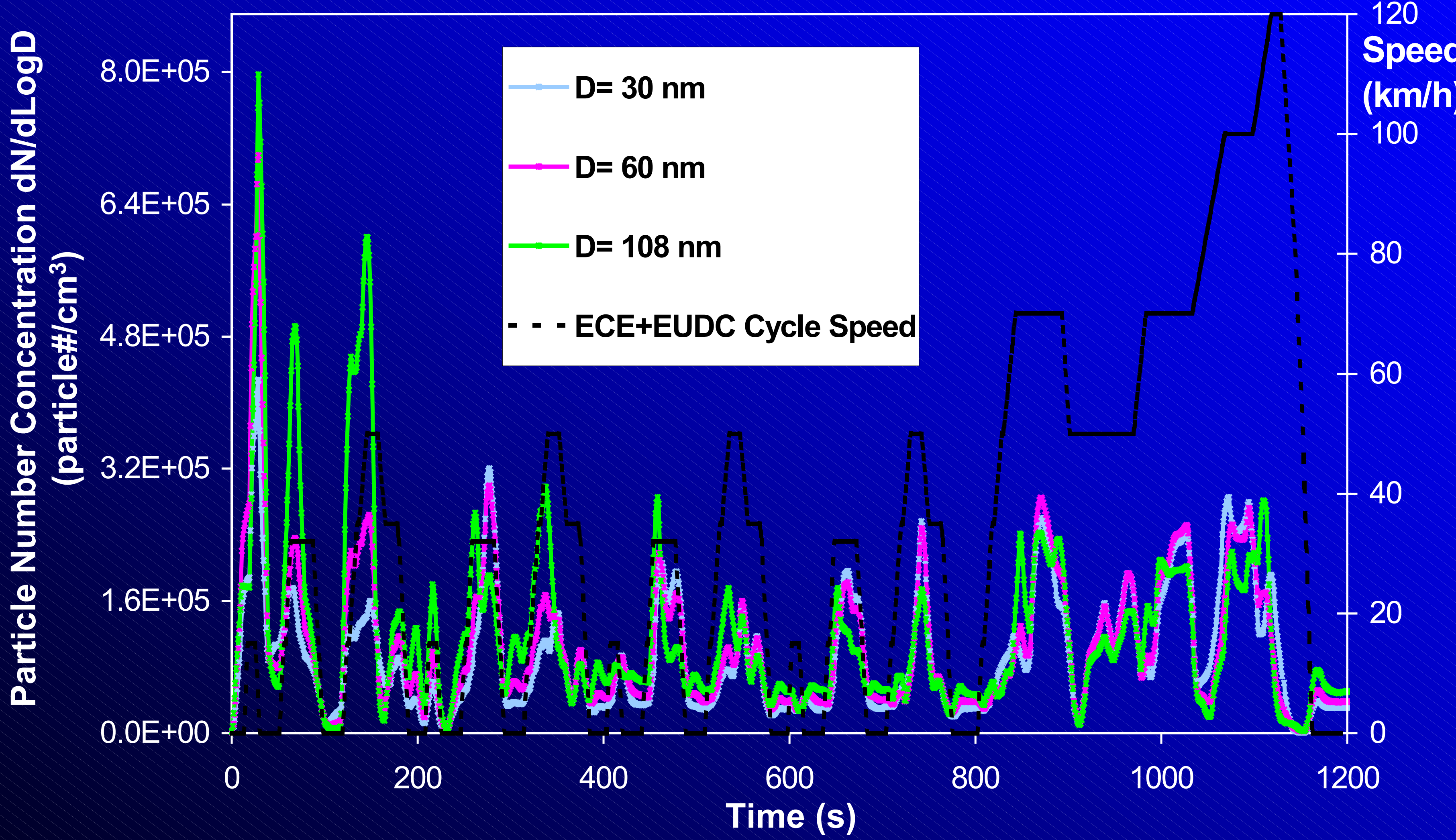
PM from a GDI-engined passenger car (Mitsubishi Carisma 1.8 litre)



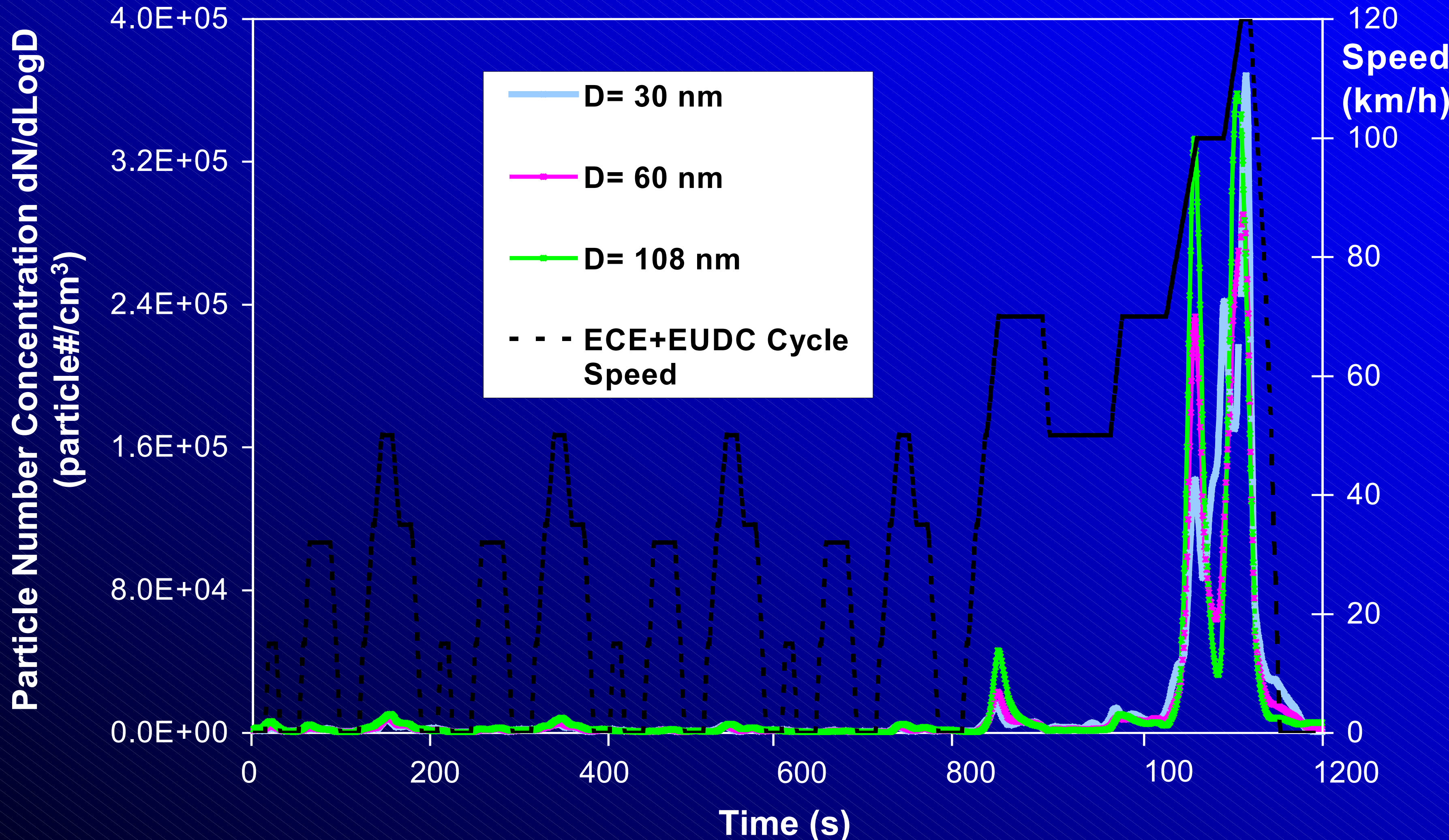
PM Simulation - Collection efficiency by number vs particle size, at 400°C and 20 °C



GDI vehicle on chassis dynamometer
No Wall-Flow Filter
European Drive Cycle (ECE + EUDC)

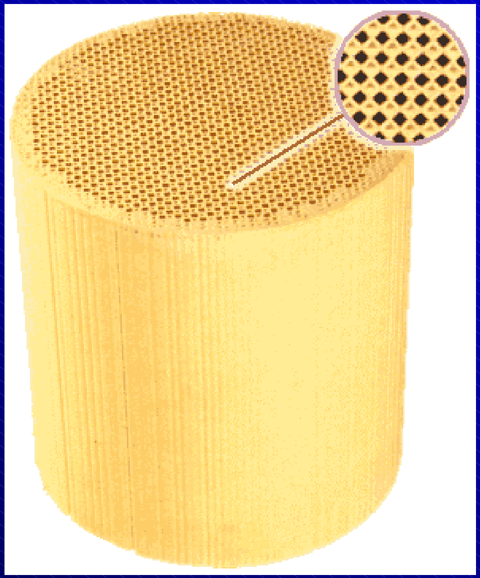


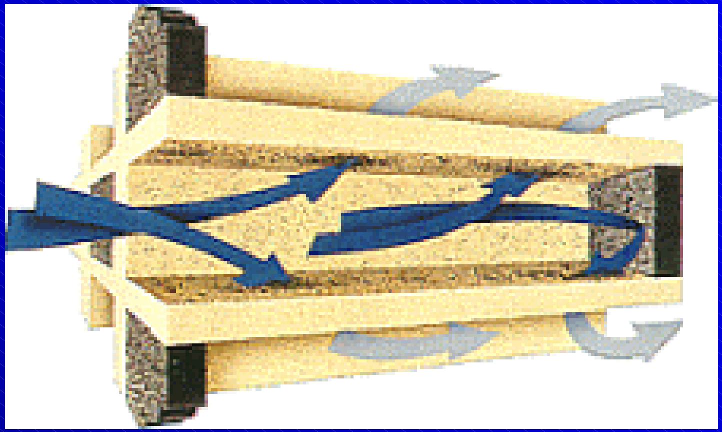
GDI vehicle on chassis dynamometer with Wall-Flow Filter European Drive Cycle (ECE + EUDC)



Wall-flow filters

- Experimental investigation of the effect of a cordierite wall-flow filter on simulated PM & GDI engine-out PM at different operating temperatures for long exposure times showed:



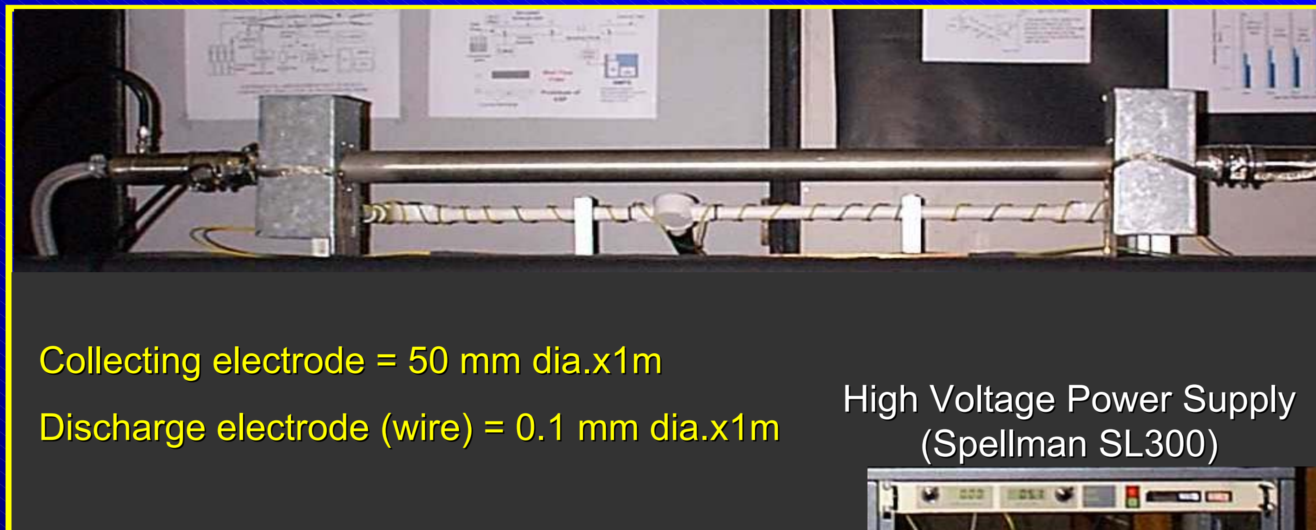


◁ High collection efficiency by number

◁ Can be operated at high temperature

◁ but…not suitable for mobile catalysts under gasoline engine exhaust conditions

Prototype Electrostatic Precipitator (ESP)



Collecting electrode

Corona glow

Catalyst for collected PM destruction under typical gasoline exhaust conditions

- Gasoline exhaust conditions:

  - High gas temperature, 600°C - 800°C / diesel 200°C - 400°C

  - Low oxygen, 300 ppm post TWC / diesel typically 12%

  - Low nitric oxide, 10 ppm post TWC / diesel up to 1000 ppm

  - High water & carbon dioxide, typically 9 -10% and 18% respectively

- Contact between catalyst and soot is crucial

Alkali metal catalyst supported on reducible metal surface with low porosity meet requirements

Integrated Catalysts / ESP Prototype

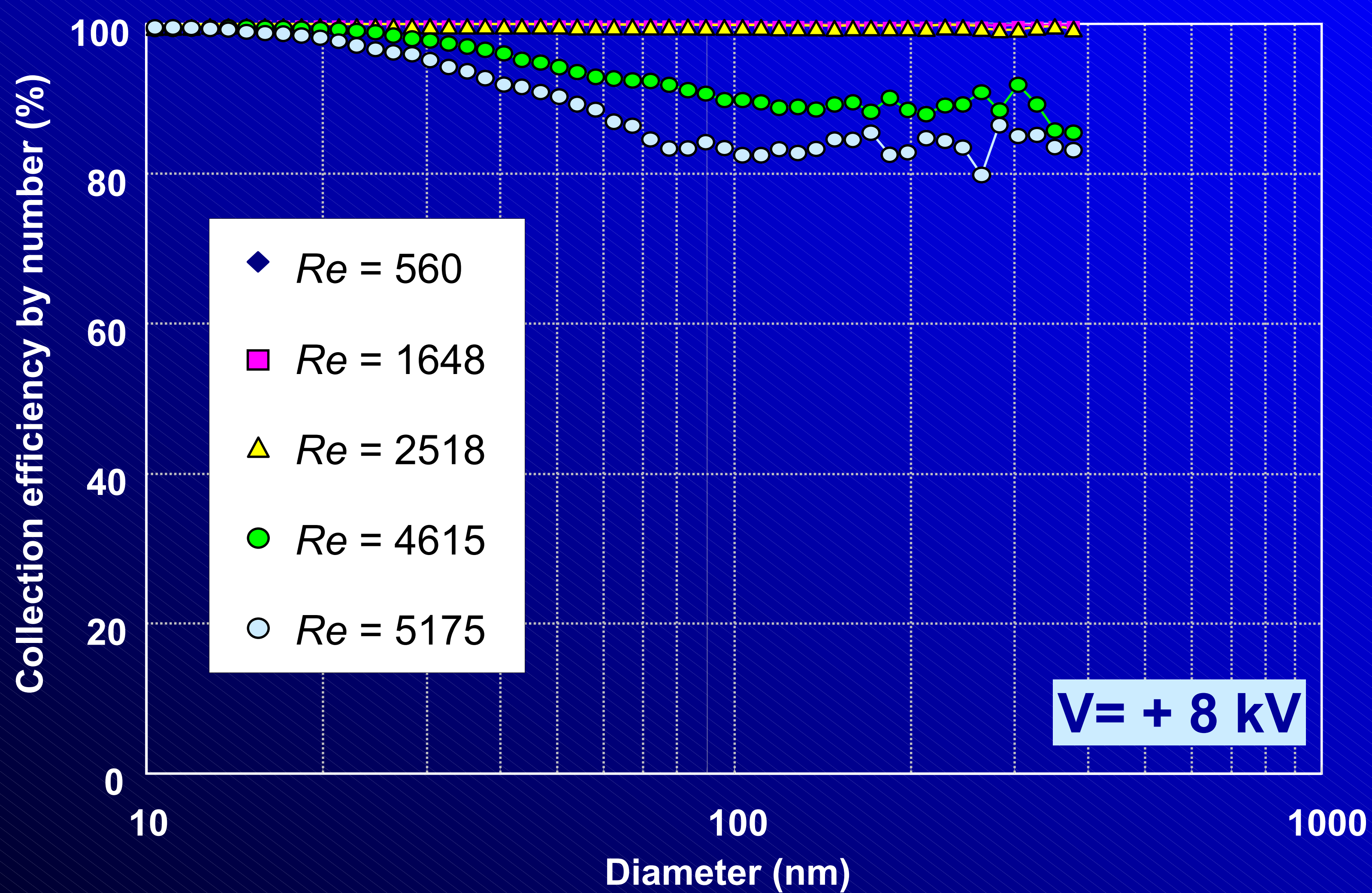
- Investigation within a laboratory PM simulation of both un-coated & alkali metal catalysts coated ESP for:

  - Different gas temperature (20 ^{0}C - 400 ^{0}C)

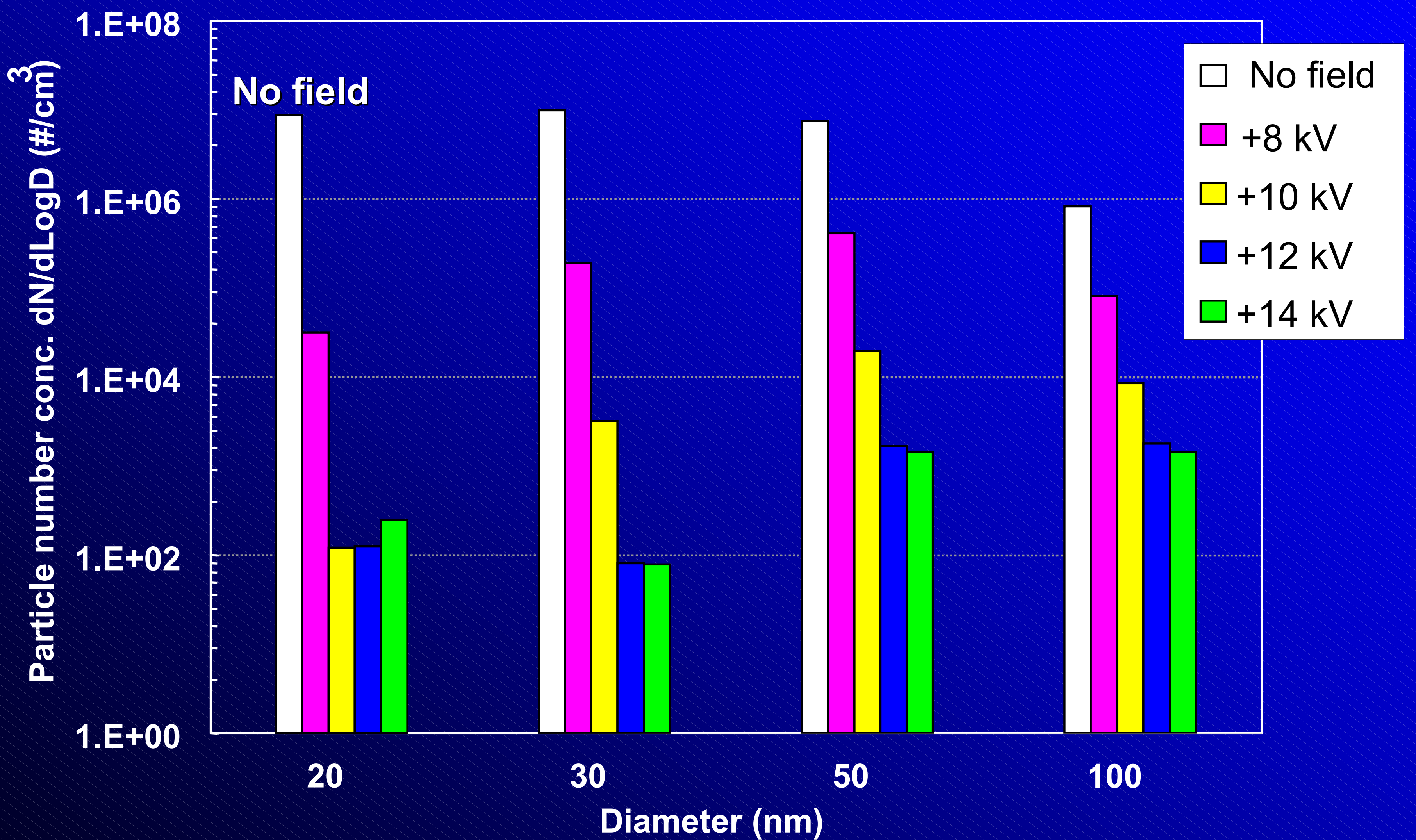
  - Different gas residence time & Reynolds number (*Re*)

  - Increasing electrode voltages up to +14 kV

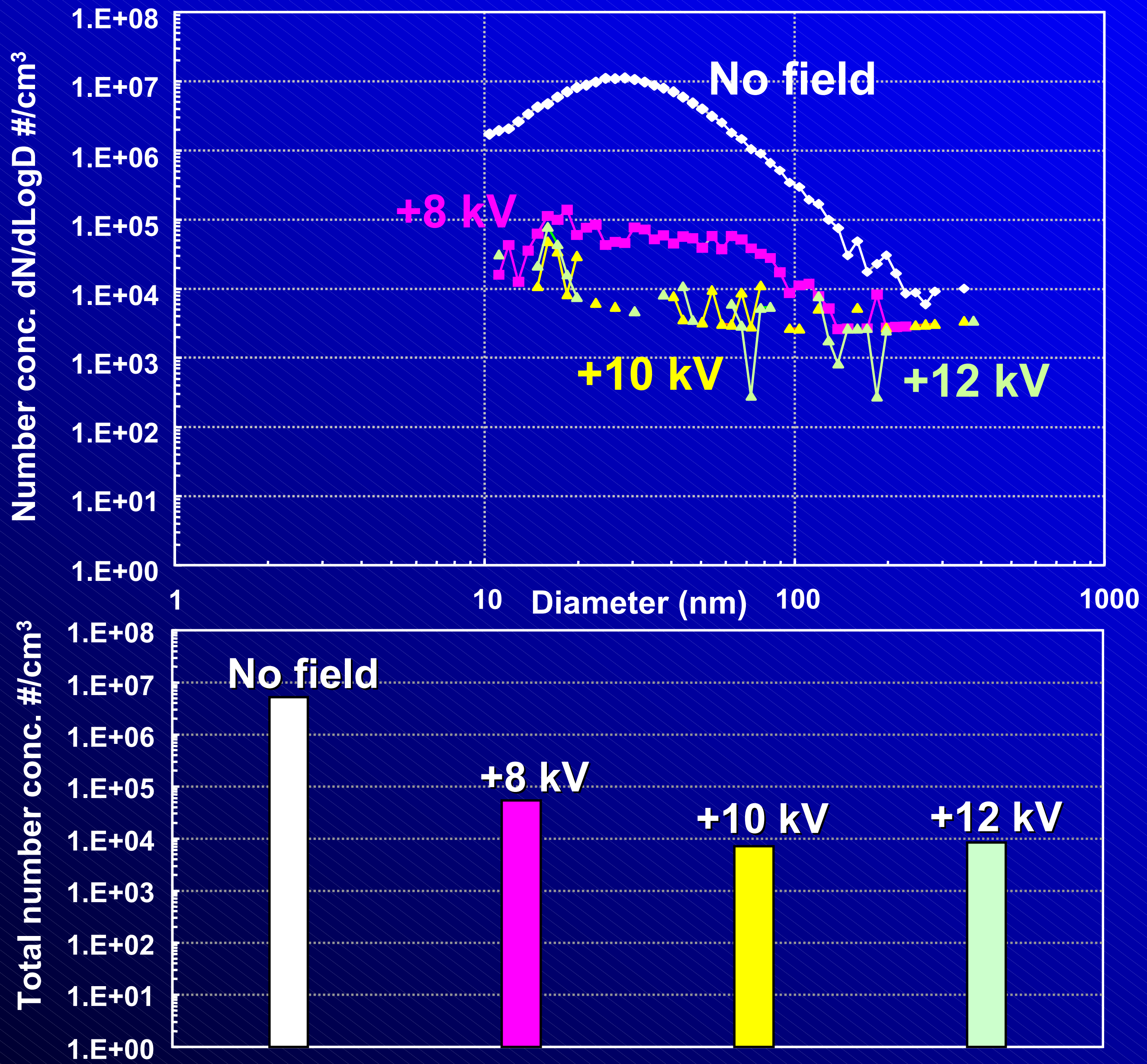
Un-coated ESP collection efficiency by number vs particle size, at different *Re* - T= 20 ^{0}C, V= + 8 kV



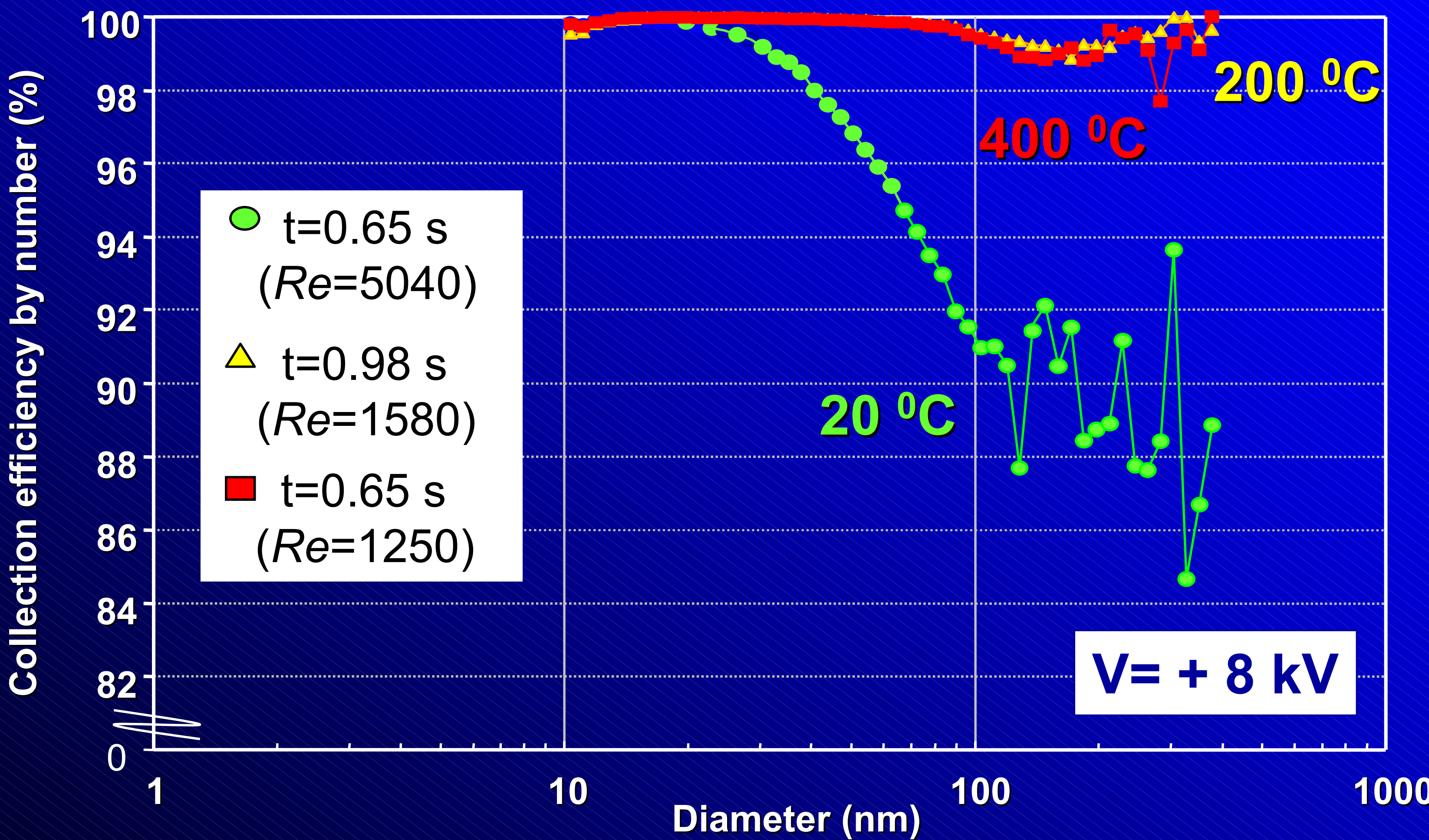
Un-coated ESP: effect of voltage on particle sizes (20, 30, 50,100 nm) at T= 20 ^{0}C *Re*= 4615, t~ 0.65 s



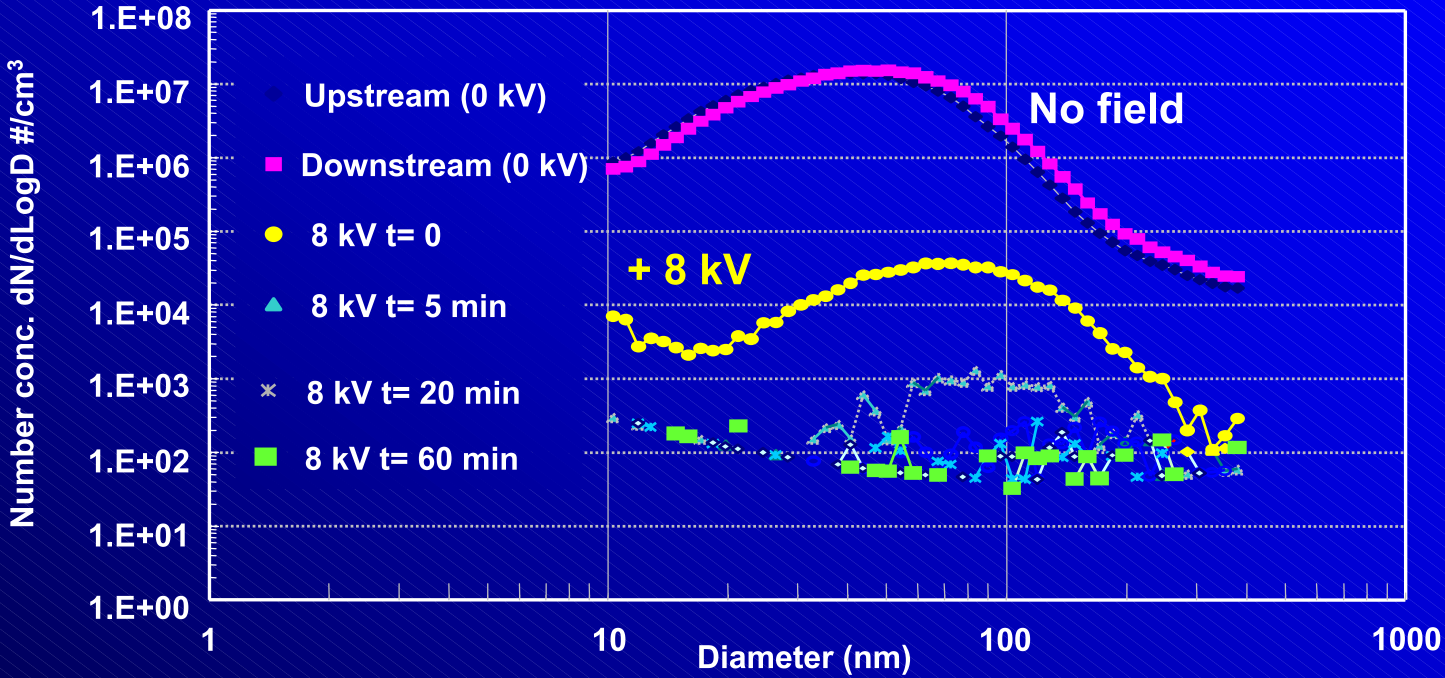
Un-coated ESP: effect of increasing voltage at Re=3408, T_{Gas}= 230 ^{0}C, gas residence time t~ 0.40 s

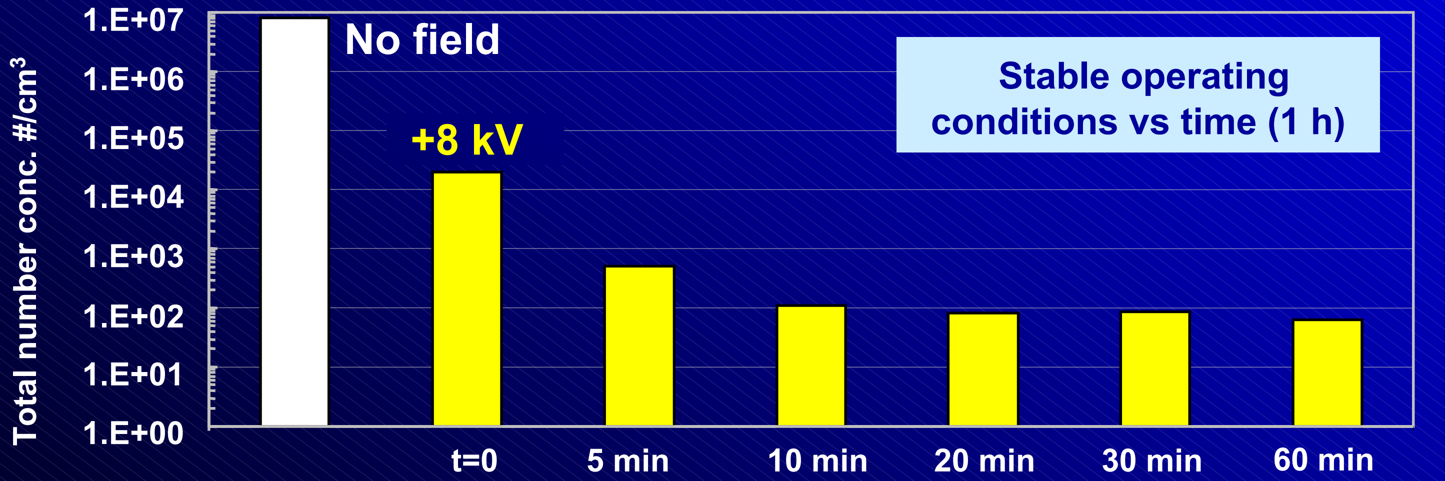


Un-coated ESP: collection efficiency by number for different T & residence time (t) at V=+ 8 kV

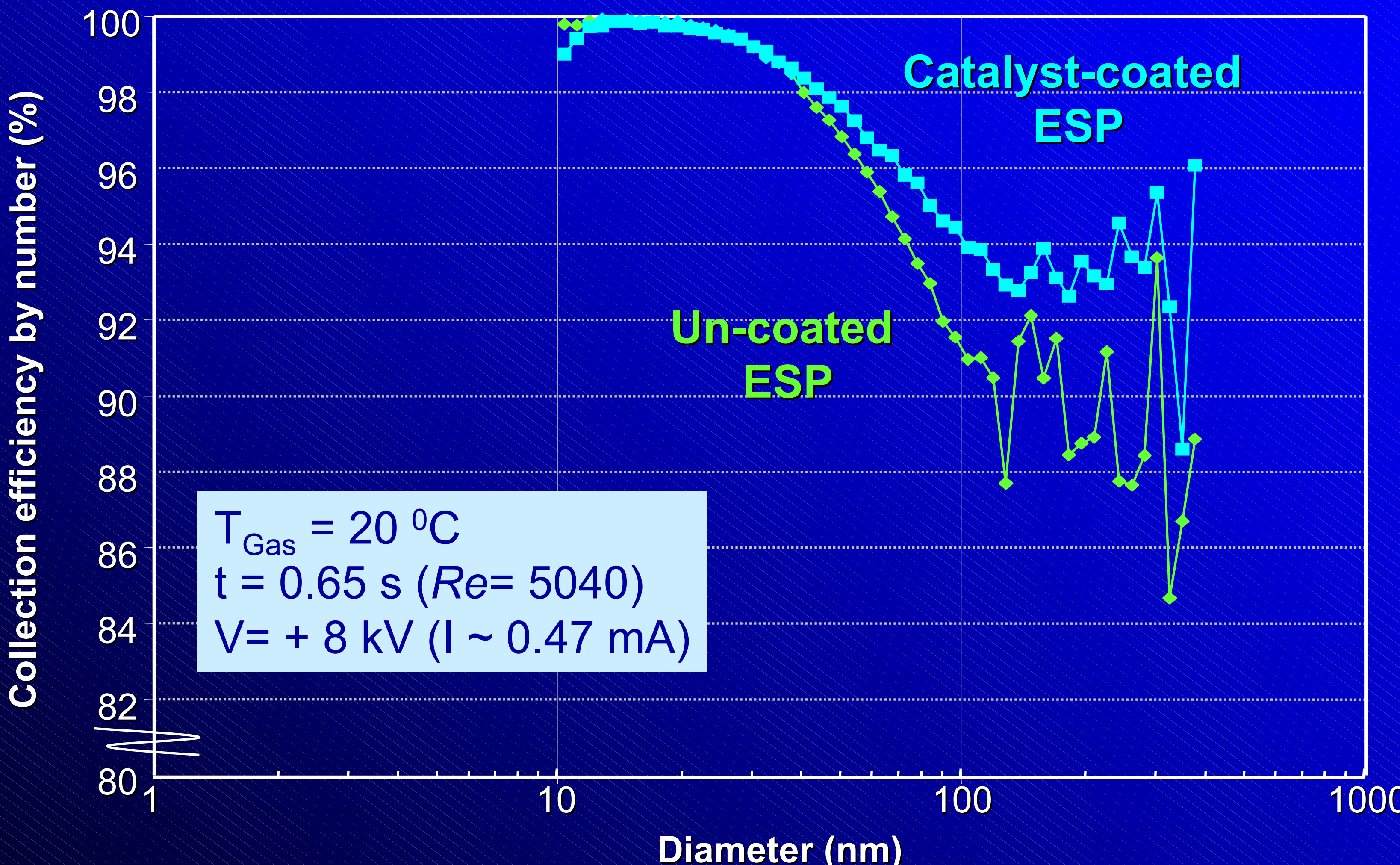


Catalyst-coated ESP: effect of + 8 kV vs time (1 h), gas residence time t ~ 0.75 s, *Re*= 1390, T= 300 ^{0}C



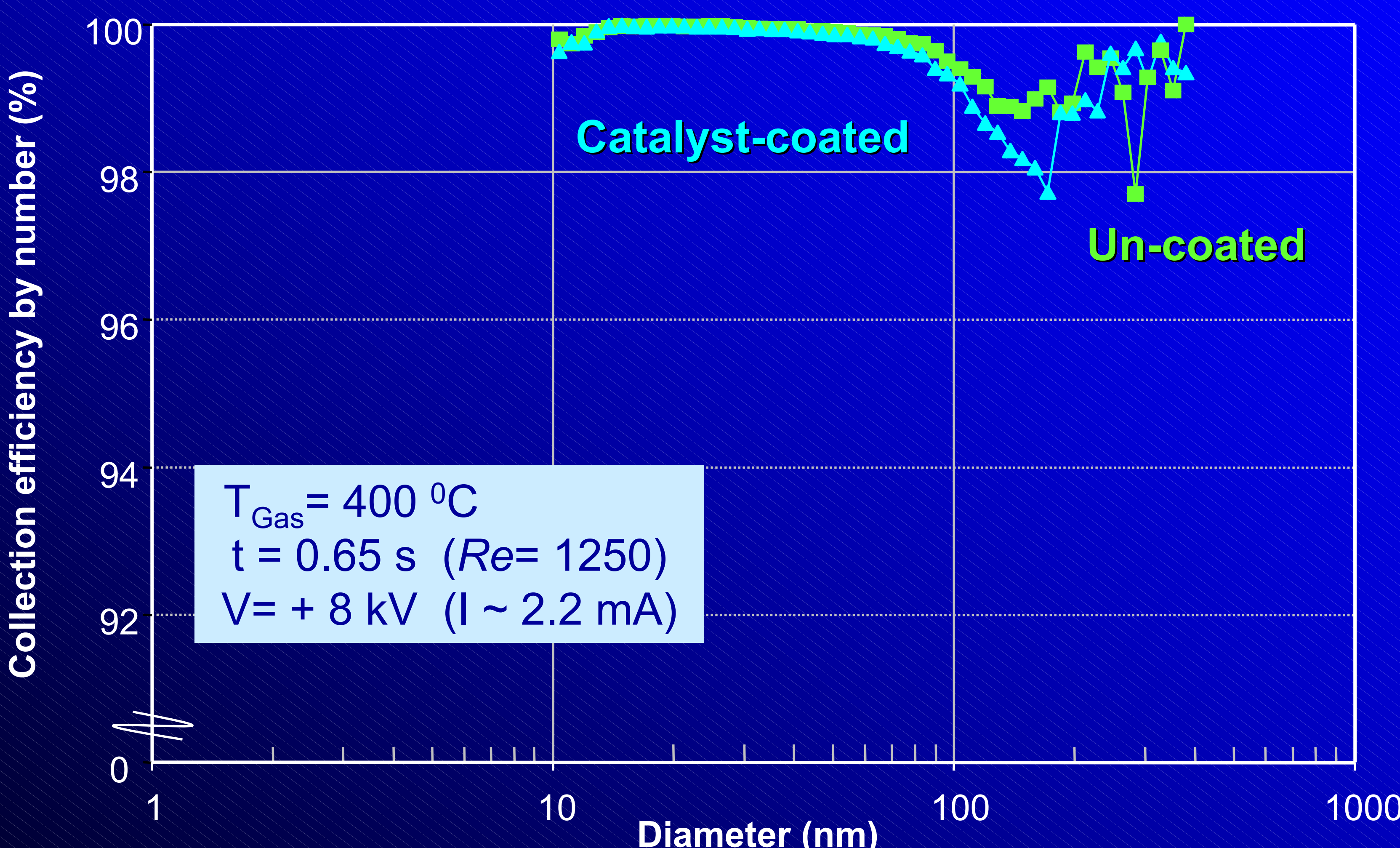


Collection efficiency by number at the ESP outlet at V= + 8 kV - T= 20 ^{0}C, Re= 5040, t= 0.65 s



- No reduction in trapping performance with catalyst coating

Collection efficiency by number at the ESP outlet at V= + 8 kV - Gas at T= 400 ^{0}C, Re= 1250, t= 0.65 s



- No reduction in trapping performance with catalyst coating

Conclusions

- Prototype Electrostatic Precipitator (ESP) more promising than Wall-flow filters for reducing particle number concentrations within a PM simulation at different gas flow conditions / temperature up to 400 ^{0}C

- Alkali metal catalysts meet requirements for destroying PM deposited in the prototype ESP under gasoline engine exhaust conditions

- At long gas residence time (t ~ 5.8 s) particle number concentrations were reduced from the baseline by a factor of ~10^2 at 20 nm and ~ 10^5 at 100 nm particle sizes applying a voltage V= + 8 kV

- At shorter gas residence time (0.4< t < 1 s) similar reductions were achievable by applying higher voltage, +10 -12 kV instead of +8 kV

- High collection efficiency by number (above 95%) was obtained with integrated catalysts/ESP within the PM simulation

Current & Future Work

- Integrated Catalysts / ESP testing is in progress to investigate the effect of:
  - ◁ Engine-out PM (DISI or GDI engine) on device performance
  - ◁ Catalyst performance under real gasoline exhaust conditions
- Modeling of nanoparticle transport and deposition in the ESP prototype is in progress to determine optimum ESP operating conditions for high efficiency particle capture
- Future modifications of the ESP design will follow the experimental investigation and modeling

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