Nano-particles production by dielectric barrier discharge

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Size-calibrated standard aerosol smaller than 100 nm are required to study the size-dependent properties of nanomaterials and to calibrate size measurement tools. Nucleation in gas phase is an economic route in that respect. In this work, a filamentary Dielectric Barrier Discharge (DBD) with low energy filaments (~ µJ) is studied to produce nano-aerosols with controlled properties (d_p < 100 nm, chemical composition, concentration). The experimental set-up is shown on figure 1.

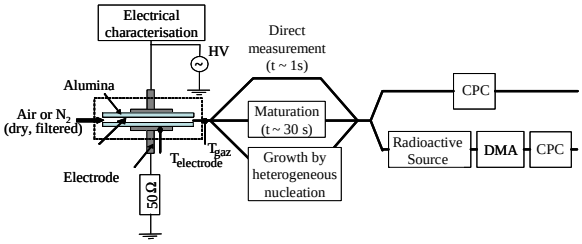

Figure 1. Experimental set-up.

The plane-to-plane DBD reactor is made of rectangular electrodes covered by 0.55 mm thick alumina plates (gap length: 0.5 or 1 mm). A dry and filtered gas (air or nitrogen) is injected in DBD. AC voltage amplitude, discharge current and electrical power are measured, as well as electrode and gas temperatures. A CPC is used for concentration measurements and a DMA or a nano-DMA for size distribution determination. Particles are studied either directly after their production by DBD or after a step of growth by heterogeneous nucleation.

Primary particles from DBD: analysis by EDX and determination of distances between atomic planes on HRTEM images have proved that the dielectric material covering electrodes represents at least a part of particles chemical composition. It shows that particles formation is due to the nucleation of vapours produced under the effect of filaments.

For short transit times (~ 1 s) low concentrations are measured with CPC (10^{-2} to 10^{-1} cm^{-3}), in air as in nitrogen. It indicates that most particles are smaller than 5 nm (smallest diameter detected by CPC). Indeed, low energy DBD filaments lead to vapours and primary particles densities preventing fast coagulation.

In N_2, when transit time is increased to 16 s, particles grow by coagulation up to a modal diameter of 3 nm. Measured concentration and modal diameter reach a maximum a few tens of minutes after DBD ignition and then decrease slowly during several hours. This still has to be explained. In air, no particle is detected after a step of maturation, even with a nano-DMA. So either particles remain smaller than the nano-DMA cut-off diameter (2 nm) or they have a diameter between 2 and 5 nm and are too diluted to be detected by this apparatus. Indeed, particles charge efficiency is weak in this range of size, even using a unipolar charger. Hence, vapour flux is lower (which is partly related to less energetic filaments) or recollection is higher in air than in N_2.

Post-DBD growth by heterogeneous nucleation: to make particles detectable by CPC (> 5 nm) and control their diameter below 100 nm, a vapour is injected in post-DBD. Its density is adjusted by tuning the temperature of an external source. In these conditions, concentrations up to 5.10^7 cm^{-3} are measured with CPC. It confirms the production of a concentrated ultra-fine aerosol by DBD.

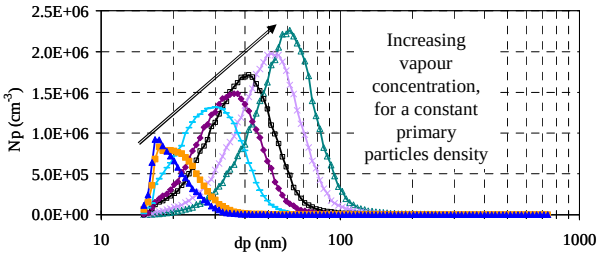

Figure 2. Size distributions of particles after post-DBD growth by heterogeneous nucleation

An example of unimodal size distributions obtained after heterogeneous nucleation is presented on figure 2. Modal diameter can be adjusted up to 60 nm, by increasing vapour density at constant primary particles concentration, which increases the amount of condensable vapour that can nucleate on each primary particle. The evolution of particles diameter cannot be due to growing coagulation since particle concentration increases with modal diameter. DBD heterogeneous nucleation can thus be used as a size-calibrated nano-particles plasma generator.

Measured particle concentration increases with diameter, keeping the same order of magnitude for modal diameters higher than 20 nm (i.e for granulometries that are not truncated due to DMA cut-off diameter). According to calculations, this phenomenon cannot be explained by an evolution in losses by diffusion, thermophoresis or electrostatic repulsion. So it is probably due to heterogeneous nucleation on smaller primary particles when saturation ratio increases. Aerosol standard geometric deviation is about 1.42. Calculations have shown that size dispersion is not related to coagulation. It could probably be reduced by optimizing the saturator geometry.